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Contents

An attempt to arrange silicates in the order of reaction energies at relatively low temperatures . . . . . John W. Gruner 137
A structural classification of fluoaluminates . . . . . A. Pabst 149
The crystal chemistry of montmorillonite . . . . . Duncan McConnell 166
Cordierite in pegmatite near Micanite, Colorado . . . . . E. Wm. Heinrich 173
Disorder in the mineral bornite, Cu_5FeS_4 . . . . . Alfred J. Frueh, Jr. 185
Twinning in the diamond . . . . . Chester B. Slawson 193
Thermal analysis studies on carbonates (I): Aragonite and calcite . . . . . George T. Faust 207
The effect of the interlayer cations on the expansion of the mica type of crystal lattice . . . . . Isaac Barshad 225
Nomographs for triclinic cell computations . . . . . W. L. Bond 239
Studies of uranium minerals (I): Parsonite and randite . . . . . Clifford Frondel 245
Studies of uranium minerals (II): Liebigite and uranothallite . . . . . Howard T. Evans, Jr. and Clifford Frondel 251
Presentation of the Roebling Medal of the Mineralogical Society of America to Herbert E. Merwin . . . . . Norman L. Bowen 255
Acceptance of the Roebling Medal of the Mineralogical Society of America . . . . . Herbert E. Merwin 258
Memorial of Frederick Noel Ashcroft . . . . . F. A. Bannister 259
Memorial of Everend Lester Bruce . . . . . J. E. Hawley 262
Proceedings of the thirtieth annual meeting of the Mineralogical Society of America at El Paso, Texas . . . . . C. S. Hurlbut, Jr. 268
Correspondents, fellows and members of the Mineralogical Society of America . . 295
Mineralogical Society (London) 320
Notes and news: Carnotite and radioactive shale in Missouri . . . . . Garrett A Muilenburg and Walter D. Keller 323
  Synthesis of uvarovite . . . . . F. A. Hummel 324
Book reviews 328
New mineral names 333

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AN ATTEMPT TO ARRANGE SILICATES IN THE ORDER OF REACTION ENERGIES AT RELATIVELY LOW TEMPERATURES*

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INTRODUCTION

There have been attempts from time to time to arrange minerals, especially the silicates, in some order which would correspond to their sequence of formation, or their order of stability, or perhaps their order of energy of reaction under a given set of conditions. Bowen's Reaction Series, familiar to all of us, is an example. Many of us who are here today were inspired by the address of our past-president, Professor M. J. Buerger (1948), two years ago on the stability relations of minerals and by his recent contribution (1948) on silica-bridging in structures. Another very creditable attempt was made by H. W. Fairbairn (1943) in his paper on packing indexes of ions. His figures, while unquestionably important, did not lead to any mineral series of either formation or stability though probably were included in both and formed a part of each of them.

It should be possible, however, to arrange the minerals in some kind of an order which would tell us something of their reaction energies under a given set of conditions, let us say for illustration, weathering or at any rate, conditions under which water plays an important role at not a very high temperature, and proton transfer becomes important.

BONDING FORCES

We know very little about the magnitude of the forces which hold ions and atoms together in minerals. Some forces are covalent or electron-sharing as in diamond and most sulfides; others are ionic as in salts and oxides. For this reason these two classes cannot be directly compared. The silicates comprise a third class in which the forces are partly ionic and partly covalent. The cations with the exception of those with very high valence like Si and Ti, and possibly Al, are tied to oxygen largely ionically. An oxide like SiO_2 on the other hand is at least 50% covalent and for this reason the atomic percentage of Si in a silicate is of paramount importance in the behavior of the mineral.

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* Address of the retiring president of the Mineralogical Society of America given at El Paso, Texas, on November 11, 1949, under the title "An Attempt to Arrange Silicates in the Order of Free Energies."

It appears then that we may have to pay heed to two proportionality factors which for our purposes may be treated separately:

(1) The factor dealing with ionic forces.

(2) The factor dealing with covalent SiO_2 forces.

				H							
				2.1							
Li	Be	B		C		N		O		F	
1.0	1.5	2.0		2.5		3.0		3.5		4.0	
Na	Mg	Al		Si		P		S		Cl	
0.9	1.2	1.5		1.8		2.1		2.5		3.0	
			/		\		\		\		\
K	Ca	Sc	Ti		Ge		As		Se		Br
0.8	1.0	1.3	1.6		1.8		2.0		2.4		2.8
Rb	Sr	Y	Zr		Sn		Sb		Te		I
0.8	1.0	1.3	1.6		1.7		1.8		2.1		2.5
Cs	Ba										
0.7	0.9										
			Fe'''		Fe'' assumed by the author						
			1.5		1.2						

FIG. 1. Electronegativities of the Elements. (After Linus Pauling)

Pauling has provided us with a means of estimating the order of magnitude of the ionic forces through his so-called Electronegativity Series which is shown in Fig. 1. He has assigned certain values to the most common ions which give their relationship to each other, that is, those farthest apart are the most electronegative with respect to each other, and have the greatest tendencies to form stable compounds with each other. In the silicates practically all cations are tied to oxygen which has a value of 3.5. We may, therefore, take some large power of the difference between 3.5 and the value of the respective cation as proportional to the bond strength. In our case, it is simpler and more expedient, as will be explained below, to take the numbers in Fig. 1 directly for our calculations, which will be illustrated by the following examples of quartz, considered here a silicate, and of orthoclase:

SiO_2, $Si=1.8$. The oxygen ions are not counted as it is assumed that all cations are tied to oxygen. As there is only one cation in SiO_2 the electronegativity is 1.8 per cation.

$KAlSi_3O_8$, K=0.8, Al=1.5, 3Si=3×1.8. Adding $0.8+1.5+(3\times1.8)=7.7$, which is the total for the formula. Dividing 7.7 by 5, the number of cations, gives 1.54 which is the electronegativity of orthoclase or microcline. If the differences between oxygen and the respective figures given in Fig. 1 had been taken, the result would have been 1.70 and 1.96, respectively, which are the complementary numbers to 1.80 and 1.54, each pair adding up to 3.50, the value for oxygen. For our purposes it is more convenient to use the numerical values 1.80 and 1.54 even though 1.54 actually stands for a greater electronegativity than 1.80, a numerical inversion to which one soon becomes accustomed.

Pauling's Table, Fig. 1, is not complete for our purposes as the element iron is left out, for the obvious reason that Fe has two possible valences. For the present purpose we may assume ferrous iron to have the value 1.2 the same as Mg, and ferric iron 1.5, like Al, though we may be certain that considerable errors are introduced by this assumption, even when as here oxidation is neglected as a factor. The value for hydrogen as given by Pauling is 2.1 which seems high to introduce into minerals containing hydroxyls or hydrogen bonds. The calculated values obtained with it appear to be of the correct order of magnitude, however.

If the silicates are arranged according to their calculated electronegativities (Table 1) it is observed that in general, the high temperature minerals of Ca and Mg are at one end and the hydrated ones are at the other. Closer examination, however, indicates plainly that these electronegativities alone do not give us the desired answer. Some important factor is still missing. It is obvious that electronegativity will not distinguish between two or more different manners of bonding of the same kinds of ions or atoms as, for example, SiO_4 framework and chain structures which are largely covalent anyway and for this reason should receive additional attention.

Such different bonding should introduce a proportionality factor in our calculations which we shall call the bridging factor, a name used first by M. J. Buerger (1948) who applied it to the manner and number of bridging between SiO_4 tetrahedra. For example, in quartz, cristobalite and tridymite all tetrahedra are directly tied to other tetrahedra of SiO_4 resulting in the highest number of bridges possible, a perfect framework, with a bridging factor, arbitrarily called 1.00. All other structures evidently will have smaller factors.

It immediately occurs to one that other atoms or ions besides Si must contribute to the bridging factor. AlO_4 tetrahedra commonly substitute

TABLE 1. ENERGY RELATIONS OF SILICATES BASED ON ELECTRONEGATIVITIES AND COORDINATIONS

Silicate	Cation Formula	Electro-Negativity	Bridging Factor	Energy Index
Quartz	Si	1.80	1.000	1.80
Montmorillonite	$H_4(Al, Mg)_{4-5}Al_2Si_6 \cdot 2H_2O$	1.80	0.948	1.77
Kaolinite	$H_8Al_4Si_4$	1.87	0.938	1.75
Pyrophyllite	$H_4Al_4Si_8$	1.80	0.962	1.73
Heulandite	$Ca_2Al_4Si_{14} \cdot 12H_2O$	1.90	0.878	1.67
Stilbite	$NaCa_2Al_5Si_{15} \cdot 15H_2O$	1.89	0.874	1.65
Lawsonite	$H_4CaAl_2Si_2$	1.78	0.928	1.65
Hydromicas	$H_4(H, K)Al_6Si_6$	1.73	0.950	1.64
Vermiculite	$H_2(Mg, Fe)_3(Si, Al)_4 \cdot 2H_2O$	1.77	0.922	1.63
Danburite	CaB_2Si_2	1.72	0.950	1.63
Euclase	$H_2Be_2Al_2Si_2$	1.73	0.938	1.62
Zircon	$ZrSi$	1.70	0.950	1.62
Bertrandite	$H_2Be_4Si_2$	1.73	0.926	1.60
Beryl	$Be_3Al_2Si_6$	1.66	0.964	1.60
Staurolite	$H_2FeAl_4Si_2$	1.69	0.940	1.59
Datolite	$HCaBSi$	1.72	0.926	1.59
Laumontite	$(Na_2, Ca)Al_2Si_4 \cdot 4H_2O$	1.80	0.876	1.58
Serpentine	$H_8Mg_6Si_4$	1.73	0.912	1.58
Chlorites	$H_8(Mg, Fe)_5Al_2Si_3$	1.73	0.912	1.58
Chloritoid	$H_4Fe_2Al_4Si_2$	1.70	0.928	1.58
Dumortierite	$HBAl_8Si_3$	1.65	0.958	1.58
Apophyllite	$KCa_4Si_8F \cdot 8H_2O$	1.82	0.862	1.57
Talc	$H_4Mg_6Si_8$	1.67	0.934	1.56
Petalite	$LiAlSi_4$	1.62	0.964	1.56
Muscovite	$H_4K_2Al_6Si_6$	1.65	0.942	1.55
Topaz	Al_2SiF_2	1.60	0.966	1.55
Tourmaline	$H_4NaFe_3B_3Al_6Si_6$	1.65	0.942	1.55
Sillimanite	Al_2Si	1.60	0.966	1.55
Kyanite	Al_2Si	1.60	0.966	1.55
Andalusite	Al_2Si	1.60	0.966	1.55
Margarite	$H_4Ca_2Al_8Si_4$	1.64	0.938	1.54
Analcime	$NaAlSi_2 \cdot H_2O$	1.70	0.898	1.53
Pumpellyite	$HCa_2Al_3Si_3 \cdot H_2O$	1.65	0.915	1.51
Cordierite	$Mg_2Al_4Si_5$	1.58	0.958	1.51
Mullite	Al_6Si_2	1.57	0.962	1.51
Natrolite	$Na_4Al_4Si_6 \cdot 4H_2O$	1.69	0.888	1.50
Axinite	$HCa_2BFeAl_2Si_4$	1.60	0.938	1.50
Albite	$NaAlSi_3$	1.56	0.956	1.49
Anthophyllite	$H_2Mg_7Si_8$	1.59	0.934	1.49
Glaucophane	$H_2Na_2Mg_3Al_2Si_8$	1.58	0.940	1.49
Prehnite	$H_2Ca_2Al_2Si_3$	1.60	0.934	1.49
Phenakite	Be_2Si	1.60	0.934	1.49

TABLE 1—(*continued*)

Silicate	Cation Formula	Electro-Negativity	Bridging Factor	Energy Index
Pollucite	$H_2(Ca, Na)_2Al_2Si_5$	1.61	0.920	1.48
Orthoclase-Microcline	$KAlSi_3$	1.54	0.954	1.47
Zoisite	$HCa_2Al_3Si_3$	1.56	0.938	1.46
Epidote	$HCa_2(Al, Fe)_3Si_3$	1.56	0.934	1.46
Lithium mica	$H_4K_2Li_3Al_5Si_6$	1.57	0.924	1.45
Tremolite	$H_2Ca_2Mg_5Si_8$	1.56	0.932	1.45
Hornblende	tremolite-pargasite	1.56	0.928	1.45
Spodumene	$LiAlSi_2$	1.53	0.946	1.45
Anorthite	$CaAl_2Si_2$	1.52	0.950	1.44
Norbergite	H_2Mg_3Si	1.60	0.900	1.44
Marialite	$Na_4Al_3Si_9Cl$	1.52	0.938	1.43
Phlogopite-Biotite	$H_2KMg_3AlSi_3$	1.55	0.916	1.42
Jadeite	$NaAlSi_2$	1.50	0.946	1.42
Pyrope	$Mg_3Al_2Si_3$	1.50	0.938	1.41
Pargasite	$H_4Na_2Ca_4Mg_9Al_4Si_{13}$	1.51	0.924	1.40
Enstatite	Mg_2Si_2	1.50	0.934	1.40
Aegerine	$NaFeSi_2$	1.50	0.934	1.40
Almandite	$Fe_3Al_2Si_3$	1.50	0.930	1.40
Meionite	$Ca_4Al_6Si_6 \cdot CO_3$	1.49	0.935	1.39
Chondrodite	$H_2Mg_5Si_2$	1.54	0.904	1.39
Leucite	$KAlSi_2$	1.47	0.942	1.38
Pectolite	$H_2Na_2Ca_4Si_6$	1.49	0.920	1.37
Vesuvianite	$H_4Ca_{10}Mg_2Al_4Si_9$	1.48	0.918	1.36
Sapphirine	Mg_2Al_5Si	1.46	0.934	1.36
Humite	$H_2Mg_7Si_3$	1.50	0.906	1.36
Titanite	$CaTiSi$	1.47	0.928	1.36
Diopside	$CaMgSi_2$	1.45	0.930	1.35
Augite	$(Ca, Mg, Fe)_2(Si, Al)_2$	1.45	0.930	1.35
Grossularite	$Ca_3Al_2Si_3$	1.43	0.934	1.34
Clinohumite	$H_2Mg_9Si_4$	1.48	0.906	1.34
Nepheline	$NaAlSi$	1.40	0.928	1.30
Wollastonite	$CaSi$	1.40	0.926	1.30
Andradite	$Ca_3Fe_2Si_3$	1.42	0.918	1.30
Forsterite	Mg_2Si	1.40	0.912	1.28
Fayalite	Fe_2Si	1.40	0.912	1.28
Kaliophilite	$KAlSi$	1.37	0.924	1.27
Gehlenite	Ca_2Al_2Si	1.36	0.920	1.25
Akermanite	Ca_2MgSi_2	1.36	0.914	1.24
Sodalite-Hauynite	$Na_4Al_3Si_3Cl$	1.35	0.904	1.22
Monticellite	$CaMgSi$	1.33	0.906	1.20
Merwinite	Ca_3MgSi_2	1.30	0.902	1.17
Cuspidine	$Ca_4Si_2F_2$	1.27	0.900	1.14
Larnite	Ca_2Si	1.27	0.900	1.14

for SiO_4. What are their values? Each atom or ion ought to contribute to the bridging factor in proportion to the strength of its bonds. This strength is expressed by the charge on the ion divided by the number of coordinate bonds reaching this ion which corresponds to Pauling's "valence bond."

The coordinate numbers or bonds of the ions are well known and need no further discussion except for Al. Al has sometimes 4, in other structures 6, coordinates. In the present study we cannot distinguish between them. On the basis of packing, hardness, melting points and other less tangible properties it may be assumed that the two are of very similar valence bond strength. If there is any choice, I would assign a greater one to Al with 6 coordinates under the lower temperature conditions. A valence bond, therefore, of 3/4 is assigned to all Al and ferric ions.*

An example may now be calculated. The sum of the valence bonds in $KAlSi_3O_8$ is $1/10+3/4+3=77/20$. This divided by the number of cations, 5, gives 0.77 a value which will be called the coordination coefficient.

If the total electronegativity of $KAlSi_3O_8$ were multiplied by this coefficient the wrong value would be obtained because this coefficient is introduced only to modify a portion, perhaps 20%, of the bonding forces contained in the electronegativity values. 80% of the electronegativity factor is used unchanged. Why are 20% and 80% respectively chosen? Because by trial and error it was found that this proportioning gives us the best empirical agreement. For $KAlSi_3O_8$ the bridging factor would be, therefore, $0.77\times0.20+0.80=.954$, as compared with unity, which was selected for quartz. This factor when multiplied by the electronegativity 1.54 results in the value 1.47, which I shall call the energy index of $KAlSi_3O_8$, that is of orthoclase-microcline.

About 90 silicates have been arranged according to their energy indexes in Table 1 and in subsequent tables containing special groups of minerals. In order to make the formulas as simple as possible, oxygen has been omitted except from water molecules. Isomorphous mixtures have been avoided as far as possible. The cations are given in the order of valence. Column 1 contains the electronegativity factor, column 2 the bridging factor. The coordination coefficients have been omitted. They may, however, be calculated from the information supplied in Table 1. The general spread between the indexes is not large but one may recall that they would have to be raised to some high, though unknown, power to give us comparable energy relations. Also the order is reversed in the tables, the highest energies having the lowest numerical values.

---

* Valence bonds of ⅛ have been assigned to Ca and Na, 1/10 to K and ½ to H.

DISCUSSION OF ARRANGEMENT

An inspection of the order of the silicates shows:

(1) The presence of hydrogen, mostly as (OH), in the formulas puts these silicates largely in the upper half of the table.

(2) A water molecule is treated like two hydrogens in the electronegativity factor but its bonds do not figure in the coordination coefficient except in the sum of the ions or atoms used as divisor because the water molecule can hardly be said to be tied strongly to the other atoms, but it does occupy much space and in this manner weakens the structure materially. Without this treatment of H_2O, particularly in zeolites, these minerals would be quite out of line with respect to the other silicates. Even so, the zeolites behave in a manner (Stevens, 1948) which suggests several kinds of H_2O molecules (Gruner, E. 1948) in their structures. A somewhat similar role seems to be played by such radicals as Cl in marialite, meionite, and sodalite, for they, too, are largely responsible for the loose packing and weakening of structures. For this reason, they appear only in the divisors of the sums of the valence bonds.

(3) Quartz or its polymorphs, tridymite and cristobalite, occupy a unique position at the head of the silicates, though tridymite and cristobalite have relatively high energies and should not be placed with quartz, for which reason they were omitted entirely. Unfortunately, our scheme does not allow the proper placing of polymorphs like kyanite, sillimanite and andalusite, unless we include such properties as Fairbairn's "Packing Index," which would permit the separation of such trimorphs, placing kyanite, for example, somewhere before and andalusite after sillimanite in our table.

(4) Beryllium silicates are higher in the table than expected on first thought, while lithium silicates, like spodumene and lithium mica, are considerably lower than expected. If one recalls that spodumene decomposes very readily and the Li ion as a whole has a very great tendency to become hydrated this difficulty disappears. Calcium boron silicates, datolite in particular, are much higher in the table than would be expected. The high electronegativity of 2.0 for B is partly responsible for these positions. Boron compounds, admittedly, are not understood very well at present.

(5) Silicates which we associate with "dry" magmas have higher energies than those of "wet" magmas. For this reason, they alter easily under the assumed conditions to the minerals at the beginning of the table, apart from the decomposition to oxides and carbonates which are not discussed here.

Many silicates have been omitted because they present such complicated isomorphous mixtures that their energy indexes would be questionable, though they might be between those of the end members. There is no question, however, that any stress caused by mixing will be expressed by higher free energy.

Geologic Application

If the table is examined as a whole its usefulness is somewhat obscured by its length; but if it is broken down into smaller units either according to similarity in composition, or according to their paragenesis very interesting relations appear. Time does not permit detailed discussions, but a few selections are tabulated.

Hydrolytic silicates

In processes in which the temperature is not too high and H_2O is one of the reactants, the silicates of Table 2 are formed, commonly by decomposition of other silicates which were formed at higher temperatures and would possess higher reaction energies. Other minerals containing the hydroxyl could have been added to this table, for example, epidote, but we rarely think of them as connected with hydrolysis. It is evident that these hydrolytic silicates are among the lowest in energy.

Table 2. Hydrolytic Products

	Energy Index		Energy Index
Quartz	1.80	Vermiculite group	1.63
Montmorillonite group	1.77	Serpentine group	1.58
Kaolinite group	1.75	Chlorites	1.58
Pyrophyllite	1.73	Talc	1.56
Hydromicas	1.64+	Muscovite	1.55
		Zeolites	1.50–1.67

Aluminum silicates connected with hydrothermal activity

The hydrolytic minerals just discussed are also included in the larger group of hydrothermal Al silicates. There are also some in this group which should be only among the pegmatite minerals, perhaps though occasionally found in veins as, for example, beryl. Table 3 is highly instructive because it shows the changes which are likely to occur with a drop in temperature in hydrothermal alterations and also in veins.

Biotite, for example, would go to vermiculite, and chlorite to hydromicas, and ultimately to the montmorillonite group. Other series would be: Feldspars to muscovite and/or to the montmorillonite group, or to kaolinite; feldspars to zeolites, to montmorillonite, or kaolinite as, for example, in Yellowstone Park. In all these series quartz would be

Table 3. Aluminum Silicates Connected with Hydrothermal Activity

	Energy Index		Energy Index
Quartz	1.80	Tourmaline	1.55
Montmorillonite group	1.77	Muscovite	1.55
Kaolinite group	1.75	Topaz	1.55
Pyrophyllite	1.73	Margarite	1.54
Heulandite	1.67	Analcime	1.53
Stilbite	1.65	Pumpellyite	1.51
Hydromicas	1.64+	Natrolite	1.50
Vermiculite group	1.63	Albite	1.49
Bertrandite	1.60	Prehnite	1.49
Beryl	1.60	Orthoclase	1.47
Chlorites	1.58	Zoisite	1.46
Laumontite	1.58	Epidote	1.46
Apophyllite	1.57	Phlogopite-Biotite	1.42

one of the end products as the mineral of lowest energy having high bond stability under these conditions. Opal may be mentioned as a likely neighbor of quartz. The positions of the zeolites are somewhat doubtful.

Granitic Pegmatite Silicates

These silicates (Table 4) being high in Al, B and Be are high in bond strength and relatively low in energy. The apparent inconsistency of the Li silicates in this respect has been pointed out above. The high coordination of Be, which is 4, and its high electronegativity tend to put euclase, bertrandite and beryl a little higher in the list than expected. Alka-

Table 4. Granitic Pegmatite Silicates

	Energy Index		Energy Index
Quartz	1.80	Sillimanite	1.55
Zircon	1.62	Andalusite	1.55
Euclase	1.62	Cordierite	1.51
Bertrandite	1.60	Albite	1.49
Beryl	1.60	Phenakite	1.49
Dumortierite	1.58	Pollucite	1.48
Petalite	1.56	Orthoclase	1.47
Muscovite	1.55	Spodumene	1.45
Topaz	1.55	Lithium mica	1.45
Tourmaline	1.55	Phlogopite-biotite	1.42

lies, like Na, which are found in the composition of beryl naturally raise the reaction energy resulting in lower stability. This statement applies to the presence of any ill-fitting ions which may have entered the crystal lattice at higher temperatures when the tolerance for isomorphous mixing was much greater than at lower temperatures.

As a whole the energy indexes have a remarkably narrow range which reflects the similarity of conditions under which these silicates form. The positions of biotite and tourmaline being far apart in the table seem to be reason for the observation that the two are mutually exclusive, for tourmaline crowds out biotite. The high index of zircon indicates that besides having high stability this mineral may very well crystallize at a late stage at relatively low temperatures from wet magmas.

TABLE 5. CONTACT METAMORPHIC SILICATES

	Energy Index		Energy Index
Danburite	1.63	Meionite	1.39
Datolite	1.59	Chondrodite	1.39
Andalusite	1.55	Pectolite	1.37
Muscovite	1.55	Humite	1.36
Cordierite	1.51	Sphene	1.36
Axinite	1.50	Vesuvianite	1.36
Albite	1.49	Diopside	1.35
Orthoclase	1.47	Augite	1.35
Zoisite	1.46	Sapphirine	1.35
Epidote	1.46	Grossularite	1.35
Tremolite	1.45	Clinohumite	1.34
Hornblende	1.45	Wollastonite	1.30
Norbergite	1.44	Andradite	1.30
Marialite	1.43	Forsterite	1.28
Phlogopite-biotite	1.42	Gehlenite	1.25
Jadeite	1.42	Cuspidine	1.14
Almandite	1.40		

Contact metamorphic silicates

The contact metamorphic silicates of Table 5 have a great range as would be expected for we include here minerals which form directly in contact with the magma, as well as many which make use of or need mineralizers to form. The presence of these volatiles for example causes wollastonite to form at as low as 400° C. to 450° C. From its position in the table this would not have been expected. It may be stressed once more that the arrangement does not represent the relative temperatures of formation, but rather the ease with which the silicates may alter to others under conditions as stated. The positions of silicates like marialite and cuspidine are not too certain for no particular allowance was made in the calculations for such an anion as F, and the value of a Cl radical is not certain as mentioned above.

DYNAMOMETAMORPHIC SILICATES

This is a very instructive group of silicates. With the exception of those very high in Al the structures are either framework, layers or double chains as may be seen in Table 6. This means high bridging factors except in serpentine, biotite, and hornblende where Mg and ferrous Fe lower this factor considerably. It is noteworthy that the only two open structures, albite and orthoclase-microcline have very high bridging factors as one should expect for alkali silicates in order to be stable under differential pressures.

TABLE 6. DYNAMOMETAMORPHIC SILICATES

	Energy Index		Energy Index
Quartz	1.80	Margarite	1.54
Pyrophyllite	1.73	Cordierite	1.51
Lawsonite	1.65	Glaucophane	1.49
Staurolite	1.59	Anthophyllite	1.49
Chlorites	1.58	Albite	1.49
Chloritoid	1.58	Orthoclase (microcline)	1.47
Serpentine	1.58	Zoisite	1.46
Talc	1.56	Epidote	1.46
Muscovite	1.55	Tremolite-hornblende	1.45
Tourmaline	1.55	Phlogopite-biotite	1.42
Sillimanite	1.55	Pyrope	1.41
Kyanite	1.55	Almandite	1.40

CONCLUSION

Similar tables may be prepared, for example, of acidic, alkali, and basic igneous rock silicates, and of Bowen's Reaction Series, all of which seem to agree with the order of energy indexes, except for the inverse positions of orthoclase and albite. If the proposition as outlined proves to be correct as a first approximation its ground-work is much simpler than could ever have been expected. Many criticisms from students of thermodynamics are anticipated.

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A STRUCTURAL CLASSIFICATION OF FLUOALUMINATES*

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Introduction

During 1948, while a guest in the laboratory of Professor Frondel, I had the opportunity to look into the classification of certain mineral fluorides to be described in the second volume of the *System of Mineralogy*. Although nearly a dozen structures of fluoaluminates and related compounds had been determined, there existed no unified summary of these structures, much less a comprehensive statement of their common features. Since it appeared that these structures must be based on AlF_6 octahedra which might be discrete units or joined in chains, sheets, frameworks, and other groupings, much as silicate structures are based on linked SiO_4 tetrahedra, it appeared worthwhile to attempt a complete summary of the AlF_6 structures and to compare and contrast the results with the general features of silicate structures.

Table 1. Fluoaluminates of Known Structure

Formula	Mineral Name	Structure Type	Investigators	Year
Na_3AlF_6	Cryolite	$J2_6$	Náray-Szabó & Sasvari	1938
K_2NaAlF_6	Elpasolite		Menzer	1928
$Li_3Na_3Al_2F_{12}$	Cryolithionite	$S1_4$ (Garnet)	Menzer	1930
Tl_2AlF_5		*	Brosset	1942
$Na_5Al_3F_{14}$	Chiolite	$K7_5$	Brosset	1938
$TlAlF_4$		$H0_8$	Brosset	1937
Na_2MgAlF_7	Weberite		Bystrom	1945
$Na_x(Al_{2-x}, Mg_x)(F,OH)_6 \cdot H_2O$	Ralstonite	$E8_1$ (pyrochlore)	Pabst	1939
AlF_3		$D0_{14}$	Ketelaar	1933
$KMgF_3$		$E2_1$ (perovskite)	van Arkel	1925

* Earlier description of this structure by Brosset in 1937 was designated Type $K3_3$ in Strukturbericht.

Search of the literature led to ten structure types, determined between 1925 and 1945, for which examples are given in Table 1. For several of these other representatives are also known. The compounds are listed in order of decreasing ratio of F to Al. One compound is included which contains no Al. The reason for this will appear shortly. Compounds on which structural data are meager or confused have been omitted. Complete structure determinations have been published for all those listed except K_2NaAlF_6, elpasolite. Menzer (1928) published only an extended abstract on this but the powder pattern of natural material for this cubic substance published by Frondel (1948) supports the space group and the main features, at any rate, of the structure proposed by Menzer. The structure type designations in the table are those of the Strukturbericht. For three substances the structure type is also designated by a mineral name since the structures are known to many by these names. The fluorides listed are not the compounds on the basis of which these types were established.

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* Address of the retiring President of the Crystallographic Society of America delivered at the annual meeting of the Society at the University of Michigan, April 7, 1949.

DESCRIPTION OF STRUCTURES

Diagrams for most of the structures listed have been published in the Strukturbericht and elsewhere but they are drawn on various projections and with differing conventions so that they are not suitable for comparison. New drawings have been prepared for each of these structures using exactly the same conventions, scale and projection for all. The projection is a 30–15 degree clinographic. Identity of scale has been maintained in the reproductions. In the originals the scale was 4 centimeters per angström unit.

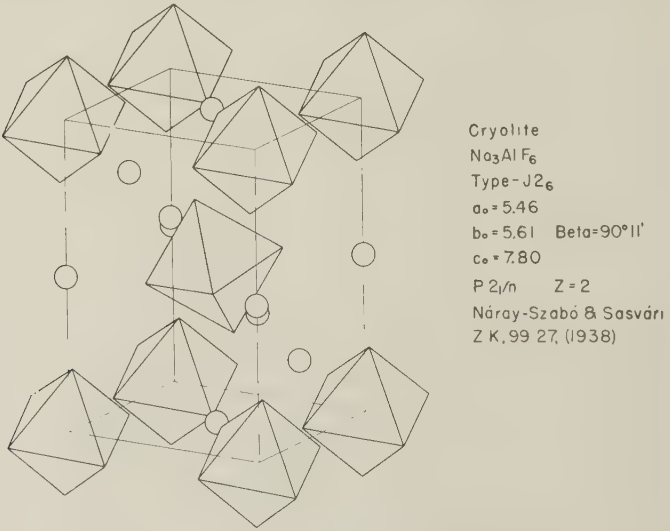

FIG. 1

The structure of cryolite was earlier unsuccessfully investigated by

Menzer. Though monoclinic the structure is pseudocubic as may be seen from the cell dimensions or from Fig. 1. Above its transition point at about 570° cryolite is isometric. The $1\bar{1}0$ plane of monoclinic cryolite corresponds to the face of a pseudocube very similar to the unit cube of elpasolite.

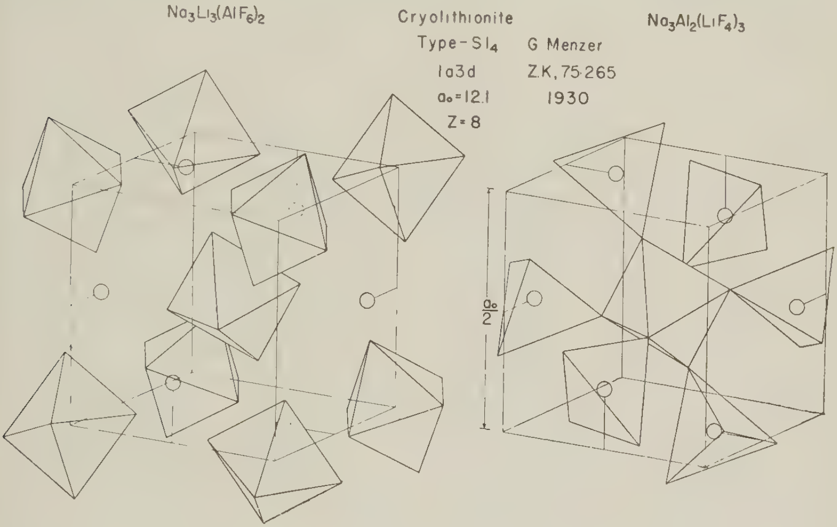

FIG. 2

For cryolithionite, which has the garnet structure, it is only necessary to draw small cubes having one eighth the volume of the unit cube to picture the structure. Two such small cubes are shown in Fig. 2. The faces of these cubes are *a* glide planes. In the left cube AlF_6 octahedra are shown and such units are bracketed in the formula written above. If we wish to group cryolithionite with the materials here considered the structure should be looked at in this way and the formula written in this manner. There is, however, an alternative way of looking at this structure which may be preferable. The small circles in the diagram represent lithiums. They are situated in each of the small cube faces. The sodiums are not shown. Each of the lithiums is at the center of a tetrahedron of fluorines and in the right hand drawing the six tetrahedra about the lithiums in each of the small cube faces are indicated. The octahedra have been omitted here except for the one in the center which has been left to show the manner in which tetrahedra and octahedra are tied together. Each fluorine is at the corner of a tetrahedron and also at the corner of an octahedron. If the formula of cryolithionite is written as at the right it resembles the formula of garnet, for instance grossularite as ordinarily written with Na taking the place of Ca and LiF_4 taking the place of SiO_4. If garnet is considered an orthosilicate then cryolithionite having precisely the same structural arrangement, should be described by the formula at the right and we may exclude it from a classification of structures based on AlF_6 groups. This is no loss since we have other examples of structures with discrete AlF_6 groups.

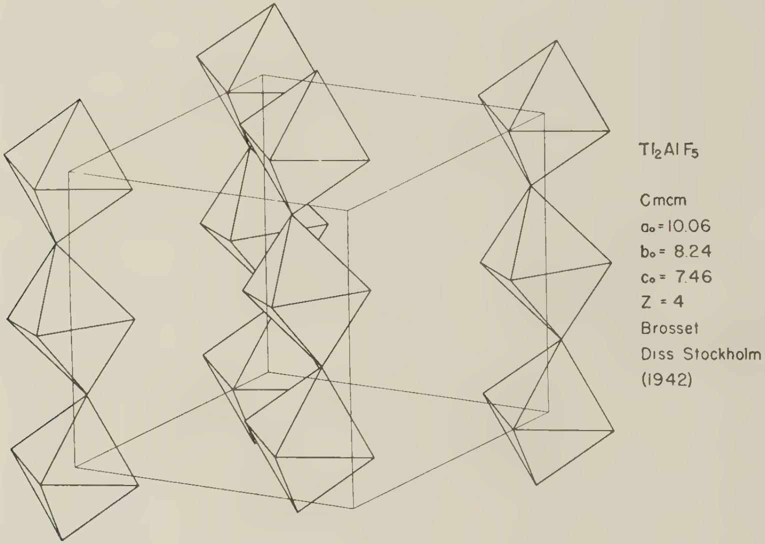

FIG. 3

If AlF_6 octahedra are linked into chains by the corners the ratio of F to Al becomes 5 and hence a chain structure might have been expected for Tl_2AlF_5. In Fig. 3, showing this structure, the light lines indicate the unit cell edges. With a base-centered lattice there is a slightly kinked chain parallel to the *c* axis through the center of the cell and at each corner. In the drawing the chain at the nearer corner is omitted. The thalliums which lie in *xyz* positions near $\frac{1}{4}\frac{1}{4}\frac{1}{4}$, etc., are not shown. The structure here pictured corresponds to that described by Brosset in his dissertation in 1942. At the same time he reported on the structure of $K_2AlF_5 \cdot H_2O$ to which he assigned the same structure with H_2O going into positions $0\frac{1}{2}\frac{1}{4}$, etc. An earlier examination of Tl_2AlF_5 had led Brosset in 1937 to assign this material to a cell of the same dimensions but to space group $C222_1$ and to propose for it a slightly different structure, which, however, also involved slightly kinked AlF_5 chains. That structure, now superseded by the one here pictured, was taken into the Strukturbericht as type $K3_3$.

The structure found for chiolite (Fig. 4) by Brosset (1938) involves sheets of linked octahedra. These octahedra are of two kinds, those with four corners shared and those with two corners shared. The latter are twice as numerous as the former. As the drawing shows this linkage results in sheets with large holes, so large, in fact, that they might accommodate another octahedron to make an unbroken sheet. Dr. R. B. Ferguson, in a communication to Professor Frondel, has referred to the chiolite sheets as "interrupted sheets." This term may be appropriate but it must be understood that the continuity of the sheets would cease only at a crystal boundary or imperfection, the word "interrupted" referring merely to the aforementioned "holes."

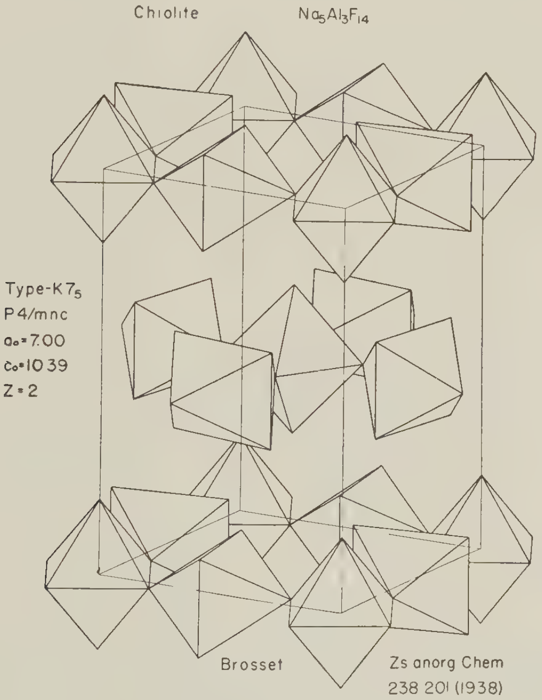

Fig. 4

A much simpler sheet structure (Fig. 5) was found in $TlAlF_4$, also studied by Brosset (1937). The light lines in the picture again indicate the edges of the tetragonal cell. There is an octahedron at the center of each cell linked to octahedra in adjoining cells making a sheet of composition AlF_4. Thalliums, not indicated in the diagram, are at cell corners and are situated between sheets that parallel the base. A segment of the next sheet above is shown in the picture at the correct height. The following year Brosset (1938) showed that $KAlF_4$, $RbAlF_4$ and NH_4AlF_4 are all isostructural with $TlAlF_4$.

In his paper on the thallium aluminum fluorides in 1937 Brosset pointed out that the structures of these compounds might be considered to belong to a series extended from cryolite involving discrete AlF_6 octahedra through chains and sheets to the network structure of AlF_3, and said that "the AlF_6 octahedron has the same sort of role in the structure of the alkalialuminumfluorides as the SiO_4 tetrahedron in the varieties of silica and the silicates." The following year, in his paper on chiolite, Brosset showed diagrammatically the relations of the three aluminofluoride structures that he had investigated. Fig. 6 shows these three structures in somewhat simplified fashion and oriented in such a manner as to show the dimensional relations. The scale of Fig. 6 and Fig. 7 is one half that used for the diagrams of single structures and the projection is 20-10 clinographic. For $TlAlF_4$ the volume outlined by the light lines is 8 unit cells, for the other two it is one cell each. Starting at the bottom with a chain structure we may think of the interrupted sheet structure as arising from it by cross linking with the turning of alternate octahedra. At the top the "holes" in the interrupted sheet are filled in and all octahedra are again in common orientation.

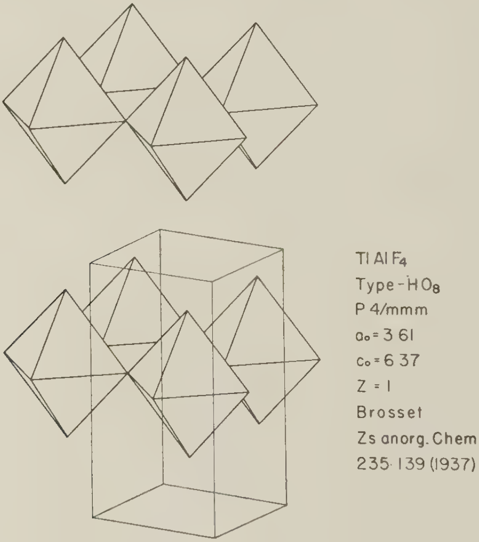

FIG. 5

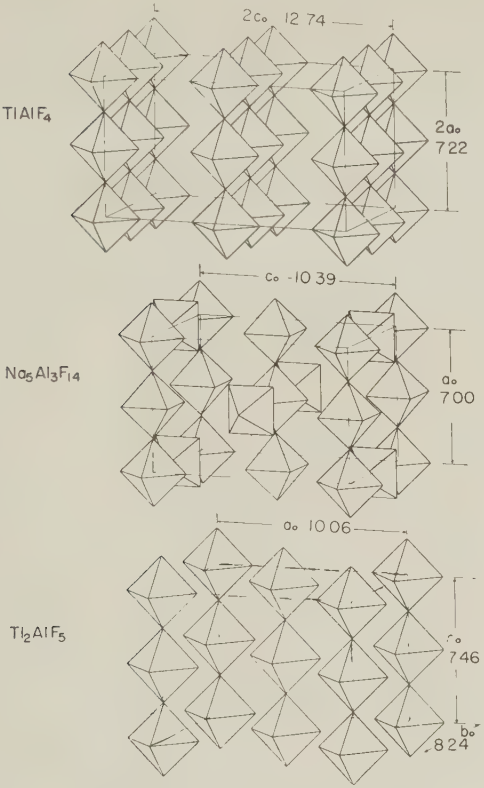

Fig. 6

Groupings not found in aluminofluorides

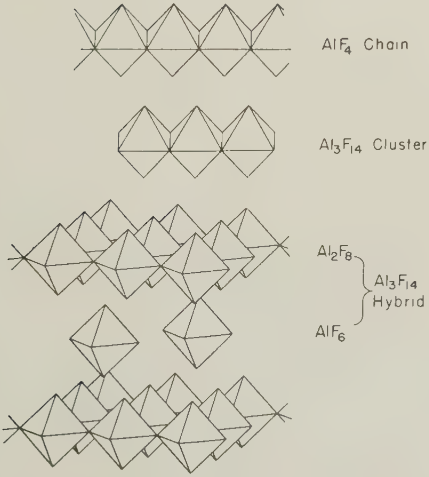

Fig. 7

In passing it is worthwhile to consider other linkages (Fig. 7) that might account for the fluorine-aluminum ratios in the structures just considered. The F/Al ratio would be 4 if octahedra were linked into endless chains by sharing of edges. No *x*-ray evidence has been found for such a situation. The ratio Al_3F_{14} would be accounted for by at least 2 other groupings besides the interrupted sheet structure. One such would be clusters of three octahedra sharing edges and making up the composition Al_3F_{14}. Another would be a hybrid structure of AlF_4 sheets, we can also call them Al_2O_8 sheets, associated with discrete AlF_6 octahedra. Brosset eliminated both of these possibilities for chiolite by intensity considerations.

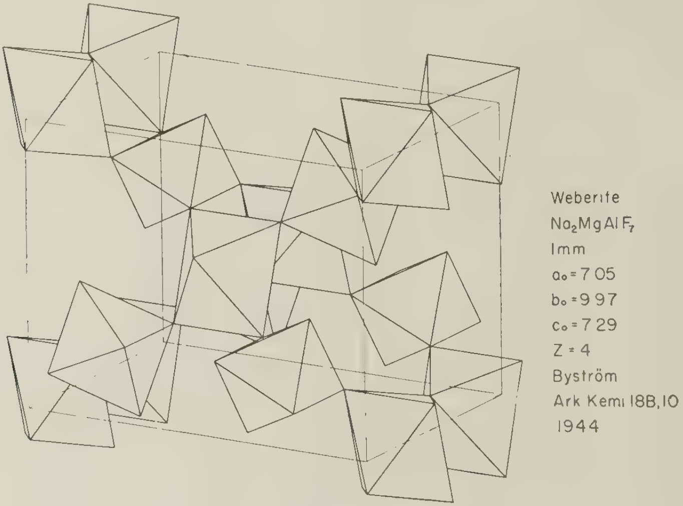

FIG. 8

In Fig. 8 the orientation chosen for weberite by Byström (1945) has been modified. He had taken the symmetry axis in the class 2*mm* as the *b* axis. By interchanging his *b* and *c* axes weberite is put in the conventional orientation for this class. The drawing shows that the structure may be looked upon as a framework of linked octahedra and that these octahedra are of two types. Half of the octahedra have one edge parallel to *b*. These share all corners, 4 with octahedra of the same kind and 2 with octahedra of the other kind. The other octahedra have one edge parallel to *a*. They each share four corners with octahedra of the first kind and have two unshared corners. Such a structure might be called an "interrupted framework" since we can go from any octahedron in the structure to any other by a path lying entirely along octahedra but there are octahedral corners at which we cannot continue to another octahedron. Byström assigns the magnesiums to the centers of octahedra sharing all corners and the aluminums to those having free corners, but points out that there is no basis in intensity considerations for checking on the distribution because of the similarity of the scattering power of Mg^{+2} and Al^{+3} and that both ions may be statistically distributed over the two positions. Byström further points out that this can be checked by a little simple arithmetic and Pauling's rules. He seems to feel that these favor statistical distribution but are not conclusive. The same author has shown (1945) that $Ca_2Sb_2O_7$ has a structure similar to weberite and here, of course, the centers of the octahedra are all occupied by like ions.

The same ratio of negative to positive ions, 7 to 2, is also characteristic of the so-called pyrochlore structure which is represented by a large number of minerals. An example is microlite, ideally $Ca_2Ta_2O_7$. The structure is cubic. Oxygen octahedra are linked at all corners to make a continuous framework of composition TaO_3 or Ta_2O_6. The extra oxygen is not involved in the linked octahedra. It has been found (Pabst, 1939) that the rare mineral ralstonite is essentially a fluoride with pyrochlore structure. The composition of ralstonite had previously been represented by a formula which seems inappropriate to me. Considered as a fluoride pyrochlore the composition may be expressed by the formula shown in Fig. 9. The octahedral corners are occupied by F or OH, the centers by Al or Mg. The drawing shows two small cubes of one eighth the volume

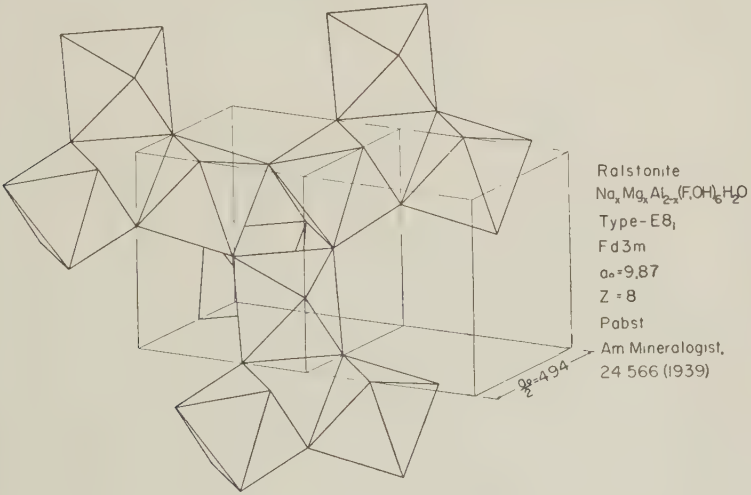

Fig. 9

of the unit cube. Their faces are *d* glide planes. Alternate small cubes contain the octahedral centers, in the drawing the one on the left. There are only as many sodium ions as there are Mg's substituting for Al's, this being required to preserve neutrality. The sodiums are in the small cube at right and so on. In the mineral the value of x is about $\frac{3}{8}$, the extra-octahedral metal positions being only partly occupied. The extra-octahedral anion positions are largely filled up with water in ralstonite. Among oxygen compounds with pyrochlore structure a few are known in which the extra anion positions are entirely unoccupied.

The possible composition range of ralstonite is unknown but it may be that some of the unnamed and unanalyzed cubic minerals from Ivigtut having slightly different refractive indices and densities are but variants of ralstonite.

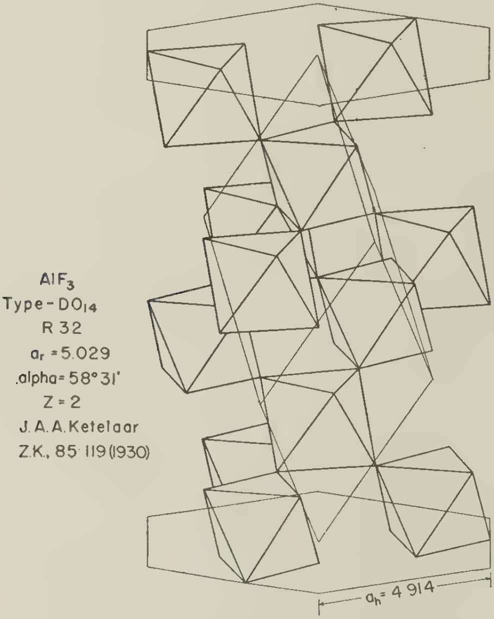

FIG. 10

In the case of weberite there remained some doubt as to whether Mg and Al are distributed statistically over equivalent positions. In ralstonite there can hardly be any doubt on this point. It may be looked upon as an AlF_3 framework structure that has received a negative charge by substitution of Mg for Al at octahedral centers, this charge being satisfied by the Na ions.

The framework structure of pure AlF_3 (Fig. 10) was investigated by Ketelaar (1933). There are also varied hydrated forms of this fluoride including the mineral fluellite, whose structures remain undetermined. AlF_3 is rhombohedral. Each cell contains two AlF_6 octahedra. These are in two orientations and each is linked at all corners to octahedra of the other orientation with centers in adjacent rhombohedral cells. To picture this linkage it is necessary to draw much more than the volume of one cell. The outline of the unit rhombohedron is shown lightly in the diagram. Above and below are shown hexagonal rings whose edges are the a_h of the hexagonal cell and which are separated by the distance c_h.

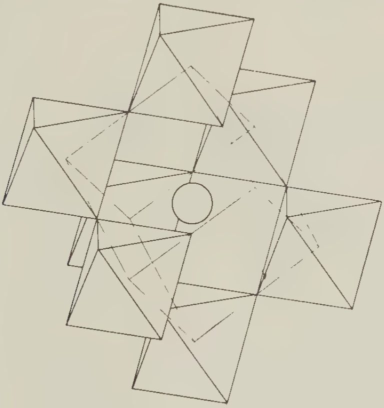
$KMgF_3$
Type-$E2_1$
Pm3m
$a_o = 4\ 0$
Z = 1
Van Arkel
Pnysica,5 162
(1925)

FIG. 11

The possibility of substituting Mg for Al at octahedral centers has been mentioned. Figure 11 shows a structure in which this substitution is carried to the limit. $KMgF_3$ has a perovskite structure (van Arkel, 1925). In the drawing the unit cube is placed with a body diagonal vertical to make it appear like a rhombohedron. Potassium, shown by the circle, occupies cube centers, magnesium cube corners with fluorine midway along the edges. The fluorines may be thought of as belonging to octahedra linked at all corners.*

In the diagram the octahedron centered on the forward cube corner has been omitted.

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* This emphasis on certain geometrical relations in $KMgF_3$ is not intended to carry any implication as to the type of bonding involved. A. F. Wells (*J. Chem. Soc. London*, 1662, 1947) has recently stated "In the perovskite structure of $KMgF_3$ no discrete complex ions exist; it is to be regarded as an infinite 3-dimensional array of K^+, Mg^{2+} and F^- ions."

SUMMARY AND GENERALIZATIONS

Table 2 shows the distances between fluorine and the ions in six-fold coordination in the substances considered. The range of Al-F distances is from 1.70 to 1.91 and for Mg-F from 1.86 to 2.00, if we consider that the larger octahedra in weberite are exclusively occupied by magnesiums. It will be remembered that this could not be established by intensity considerations. The letter *s* in the table indicates that the distance is to a shared octahedral corner. The data are insufficient to show clearly a consistent difference between these distances and the distances to unshared corners.

The last column of Table 2 shows the near constancy of the distance between the points of two contiguous octahedra in chains and sheets. This distance determines the lattice dimensions of a number of compounds in the same way in which the distance 5.2, twice the edge of a SiO_4 tetrahedron, determines the dimensions of silicate structures, e.g. the *c* axes of amphiboles and pyroxenes or the *a* axes of micas.

It would require a full tabulation of *F-F* distances to survey the departure of the fluorine octahedra in these structures from the ideal shape but the diagrams show that this departure is slight in most cases.

Seven distinct structure types have been found among the aluminofluorides. They are listed in Table 3. Of structures with separate octahedra there are several variants in addition to the one given as example. These include elpasolite and the so-called potassium cryolite and ammonium cryolite whose structures were not pictured.

For the chain structures there is also at least one more example, $K_2AlF_5 \cdot H_2O$, which is isostructural with Tl_2AlF_5. The simple sheet structure of $TlAlF_4$ exists also in the corresponding potassium, rubidium and ammonium compounds. To date the chiolite, weberite and ralstonite structures have not been found among other fluorides but the latter two are represented among oxide compounds.

From this survey it is possible to draw some general conclusions about fluoaluminate structures and to contrast them with the structures of silicates. As suggested by Brosset in 1937 the structures of these compounds are controlled by the linking of octahedra. Seven types have been recognized:—separate octahedra, chains, interrupted sheets, sheets, interrupted frameworks, and two distinctly different types of frameworks. Through this series the ratio of fluorines to six-coordinated cations decreases from 6 to 3 as the number of shared corners per octahedron increases from null to six. This is roughly comparable to the linkage of tetrahedra in silicates and the attendant change in the O/Si ratio. As shown in the diagrams, only corners, not edges, are shared the same as in the linkage of SiO_4 tetrahedra.

TABLE 2. DISTANCES IN FLUOALUMINATE STRUCTURES

Name	Formula	Distance from center to corner of octahedra	Distance between points of two contiguous octahedra in chains or sheets	
Cryolite	Na_3AlF_6	2 at 1.79 2 at 1.82 2 at 1.83		
Cryolithionite	$Li_3Na_3(AlF_6)_2$	6 at 1.81		
	Tl_2AlF_5	2 at 1.81 2 at 1.91 *s* 2 at 1.91	c_0	7.46
	$K_2AlF_5 \cdot H_2O$	2 at 1.75* 2 at 1.87 *s* 2 at 1.88*	c_0	7.45
Chiolite	$Na_5Al_3F_{14}$	2 at 1.92 *s* 4 at 1.82 *s* 2 at 1.82 4 at 1.94		
	$TlAlF_4$	2 at 1.8 *s* 4 at 1.8	$2a_0$	7.22
	$KAlF_4$		$2a_0$	7.10
	NH_4AlF_4		$2a_0$	7.18
	$RbAlF_4$		$2a_0$	7.24
	AlF_3	*s* 3 at 1.70 *s* 3 at 1.89		
Weberite	Na_2MgAlF_7	*s* 4 at 1.83 2 at 1.84 *s* 4 at 1.94 *s* 2 at 1.94		
Ralstonite	$Na_x(Al_{2-x}Mg_x)(F,OH) \cdot H_2O$	*s* 6 at 1.86		
	$KMgF_3$	*s* 6 at 2.00		

* As given by Brosset (1942). Calculation from his parameters and cell dimensions yields the values 1.65 and 1.90.

Table 3. Types of Fluoaluminate Structures

Structure type	Shared octahedral corners	F/W ratio	Example	
			Formula	Name
Separate octahedra	0	6	Na_3AlF_6	Cryolite
Chains	2	5	Tl_2AlF_5	
Interrupted sheets	2 & 4	$4\frac{2}{3}$	$Na_5Al_3F_{14}$	Chiolite
Sheets	4	4	$TlAlF_4$	
Interrupted frameworks	4 & 6	$3\frac{1}{2}$	Na_2MgAlF_7	Weberite
Frameworks	6	3	AlF_3	
Frameworks (pyrochlore type)*	6	3*		Ralstonite

* With the possibility of additional anions and an F/W ratio of $3\frac{1}{2}$.

In some respects there are notable differences from the silicates. No cases of the sharing of 1, 3 or 5 corners of AlF_6 octahedra have been found. Even numbers of corners only are shared and a close inspection of structures will show that the diametrically opposite corners are always shared together. In tetrahedra there are, of course, no diametrically opposite corners and known silicate structures involve the sharing of either 0, 1, 2, 3 or 4 corners.

In Table 4 a list of most of the recognized silicate types is arranged alongside a list of the fluoaluminate types for comparison. For instance, silicate structures include finite groups of linked tetrahedra such as Si_2O_7 pairs or Si_3O_9 and Si_6O_{18} rings. Nothing comparable, such as, say, an Al_2F_{11} pair, has been found among fluoaluminates.

Both the sheet structure with the F/Al ratio 4 and the interrupted sheet structure with the ratio $4\frac{2}{3}$ have unshared octahedral corners on both sides of the sheets. This is in marked contrast to the most familiar of the silicate sheet structures in which the unshared tetrahedral corners are all on one side though complex silicate sheets like that in the tetragonal gillespite, $BaFeSi_4O_{10}$, with unshared corners on both sides have been reported.

In the presence of very small cations RF_4 coordinations may occur together with RF_6. Among the fluoaluminates so far studied Li^{+1} is the only suitable cation. Of course, tetrahedral groups of F are well known among compounds other than the aluminofluorides, for instance, in BeF_2 $(NH_4)_2BeF_4$, $(NH_4)BF_4$, and others.

Mg is six coordinated by F in fluorides and may substitute for Al in random fashion. Such substitution has been found in ralstonite and probably exists in weberite. It also seems not unlikely that homogeneous phases intermediate between AlF_3 and, say, $KMgF_3$ could be obtained

TABLE 4. ANALOGIES BETWEEN STRUCTURES BASED ON AlF_6 OCTAHEDRA AND SiO_4 TETRAHEDRA

Type	Fluoaluminates		Silicates	
	Group Formula	Example	Group Formula	Example
Isolated octahedra or tetrahedra	$(AlF_6)^{-3}$	Cryolite Na_3AlF_6	$(SiO_4)^{-4}$	Forsterite Mg_2SiO_4
Pairs			$(Si_2O_7)^{-6}$	Thortveitite $(Sc,Y)_2Si_2O_7$
Rings			$(Si_3O_9)^{-6}$	Benitoite $BaTiSi_3O_9$
Simple chains	$(AlF_5)^{-2}$	Tl_2AlF_5	$(SiO_3)^{-2}$	Enstatite $MgSiO_3$
Bands			$(Si_4O_{11})^{-6}$	Tremolite $(OH)_2Ca_2Mg_5(Si_4O_{11})_2$
"Interrupted" sheets	$(Al_3F_{14})^{-5}$	Chiolite $Na_5Al_3F_{14}$		
Simple sheets	$(AlF_4)^{-1}$	$TlAlF_4$	$(Si_2O_5)^{-2}$	Gillespite* $BaFe(Si_2O_5)_2$
"Interrupted" frameworks	$(MgAlF_7)^{-2}$	Weberite Na_2MgAlF_7		
Pyrochlore frameworks	$((Mg_2F_6)F)^{-5}$†	Ralstonite		
Complete frameworks	AlF_3	AlF_3	SiO_2	Quartz SiO_2

* This obscure example is used because other sheet silicates all contain anions in addition to those that form part of the linked tetrahedra.

† This group formula results if x is made 2 in the ralstonite formula and the possible anion positions are entirely occupied by F.

in which such random substitution could be shown. The substitution so far has been found only in framework structures and it seems most likely that it is limited to structures in which there is a high measure of linking of octahedra. This is closely analogous to the substitution of Al for Si which is typical in silicate frameworks where many cases of halfway substitution, as in anorthite, are known. In silicate sheets halfway substitution or even beyond also occurs but the most common case is one quarter substitution as in muscovite. In the double chains or bands of amphiboles the substitution is less extensive and in the simple pyroxene chains even more restricted. It may be wholly lacking in orthosilicates though the slight excess of alumina found in some garnets might be accounted for in this way. The experimental hindrances to determination of Mg-Al distribution, referred to earlier, have their counterpart in the similar difficulty in the establishment of Al-Si distributions which has contributed to the troubles in dealing with the all important feldspars.

Fluorine octahedra centered on a small cation appear in the fluosilicates, which are known in two or more structures, as well as in fluozirconates, fluostannates and so on. In all these there are separate octahedra and they correspond roughly to the fluoaluminate structures in which no octahedral corners are shared. So far as I am aware no chain, sheet or framework structures with ions of higher charge than aluminum at the centers of the fluorine octahedra have been described.

CLASSIFICATION OF MINERAL FLUOALUMINATES

A dozen years ago the classification of silicates was considered so well established that Berman undertook a structural classification of all silicate minerals on the basis of composition and properties even in the absence of experimental knowledge of the structures. It might seem that the same could be tried for the fluoaluminates. Among the examples discussed were five minerals, all but elpasolite being from Ivigtut in Greenland. At this locality there are known at least 4 more fluoaluminates and others occur elsewhere. Fitting these into a classification seems too hazardous for two reasons. First, the F/Al ratio alone is no good guide to structure type. For instance, the weberite formula might be explained on the basis of a pyrochlore type of structure and this was, in fact, tried by Byström in working on this mineral. Second, the correlation of structure with properties of fluoaluminates is as yet very uncertain. Most of the pertinent data are given in Table 5. It may be seen

TABLE 5. PROPERTIES AND STRUCTURES OF FLUOALUMINATES

Type	Name	Composition	Symmetry	Optics	Habit	Cleavage
Separate octahedra	Cryolite	Na_3AlF_6	*P*2₁/*n*	pos.* 2V=60° B.=0.01	Pseudotet. equidimen.	poor or absent
Separate octahedra	Cryolithionite	$Li_3Na_3Al_2F_{12}$	*Ia*3*d*	Isotropic	{110} {211}†	{110} fair
Chains		Tl_2AlF_5	*Cmcm*	———	"well developed needles"	———
Interrupted sheets	Chiolite	$Na_5Al_3F_{14}$	*P*4/*mnc*	neg. uniax. B.=.007	granular massive	{001} perfect
Sheets		$TlAlF_4$	*P*4/*mmm*	———	"brittle plates"	———
Interrupted framework	Weberite	Na_2MgAlF_7	*Imm* (pseudotet.)	Nc 1.346‡ Na 1.348 Nb 1.350	no crystals	{011} poor {001}?
Pyrochlore framework	Ralstonite	$Na_x(Al_{2-x}Mg_x)(F, OH)_6 \cdot H_2O$	*Fd*3*m*	Isotropic	Octahedral	{111} imperfect
Framework		AlF_3	*R*32	pos. uniax. B. low	Pseudocubic Rhombohedrons	———

* $X = b$, $Z \wedge c = -44°$.

† The Donnay-Harker rules would require {211} and {110} as dominant forms.

‡ Optical orientation and cleavage from Ferguson (p.c. to Frondel, 4/27/48), but changed to conventional setting.

that some of the materials show properties to be expected from their structures but the data are entirely too fragmentary. This is in part due to the different techniques used by different investigators. For some materials complete structure determinations have been reported without a word about physical properties except the density. For other materials, including some of the minerals referred to a few moments ago, data on crystal habit, cleavage, density and optical properties may be known though no attempt has been made to determine the crystal structure.

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THE CRYSTAL CHEMISTRY OF MONTMORILLONITE

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Abstract

A modification of the Hofmann-Endell-Wilm structure is proposed in order to account for the thermochemical properties of montmorillonite. According to the new hypothesis, hydroxyl ions can occur in the tetrahedral layers as statistical substitutions in a manner analogous to the substitution of $(OH)_4$ for SiO_4 in the garnetoids.

The second high-temperature endotherm is interpreted as evidence of the occurrence of hydroxyls in tetrahedral configuration, whereas the hydroxyl water given off during the first high-temperature endotherm is liberated from the octahedral layer. If the silicon content of the structural unit decreases in the manner postulated, the amount of aluminum in tetrahedral coordination cannot be calculated without a knowledge of the hydroxyl content of the tetrahedral layers. This concept may explain unsuccessful calculations of the base-exchange capacity from analytical data.

Preliminary measurements on liberation of high-temperature water tend to substantiate certain portions of the hypothesis, but further data will be required in order to obtain confirmation. Sufficient reliable data probably are not available to permit rigorous calculation of structural formulas.

Introduction

Existing hypothetical structures of montmorillonite, as proposed by Hofmann, Endell and Wilm (1) and by Edelman and Favejee (2), have proven extremely useful in permitting further consideration of the isomorphism and crystal chemistry of montmorillonite and its close relative beidellite.* That both of these structures are somewhat imperfect, particularly when called upon to explain some of the observed data on clays of the montmorillonite-beidellite group, has been recognized by numerous investigators. It is the purpose of this paper to present a brief description of a new hypothetical structure for montmorillonite which, although different from either of these structural hypotheses, has some of the more attractive aspects of both.

One of the earlier difficulties that was visualized for the Hofmann-Endell-Wilm structure was the manner of adsorption of interstitial water. This difficulty has been largely overcome through the explanation offered by Hendricks and Jefferson (5). However, other incompatibilities exist between the experimental data and the Hofmann-Endell-Wilm structure, as mentioned by Grim (6, p. 241). Some of these discrepancies are more readily explained on the basis of the Edelman-Favejee structure but, here again, difficulties arise and a more adequate theoretical explanation is lacking. In the sections to follow, certain types of experimental data will be discussed and tentative explanations of these phenomena will be based on a new structural hypothesis.

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* The term beidellite is used here in the same sense as in the works of Marshall (3) and Ross and Hendricks (4), i.e., for montmorillonite in which a significant amount of aluminum occurs in the tetrahedral layer.

The interpretations furnished here will not be substantiated by detailed considerations of data, but it is believed that the hypothesis will, nevertheless, prove useful as a starting point for further discussion at a later date of the crystal chemistry of montmorillonite and related substances. It should be emphasized, therefore, that this is a brief, preliminary statement of the hypothesis and its ultimate acceptability will depend upon further consideration of numerous different types of data.

ISOMORPHISM AND BASE EXCHANGE

The present hypothesis is predicated upon two statistical observations obtained from the numerous analytical data recorded in the literature, which have been excellently collated by Ross and Hendricks (4). These observations, stated succinctly are: (1) No regular increase in the base-exchange capacity accompanies replacement of silicon by aluminum (Si→Al) in tetrahedral coordination, even after substitution of Mg in the octahedral layer has been taken into account, and; (2) according to the calculations of Ross and Hendricks, there is an apparent increase in the amount of hydroxyl water with substitution of aluminum for silicon in tetrahedral coordination, although this increase is not regular.

If the existing structures were adequate to account for these conditions, a regular increase in the amount of base exchange occasioned by $Si^{+4} \rightarrow Al^{+3}$ would be expected, provided the integrity of normal oxygen and hydroxyl ions were maintained, because one additional unsatisfied negative charge would accompany each aluminum ion introduced in tetrahedral configuration. The available analyses give no straightforward indication of such a relationship.

With regard to the second premise above, existing structures make no provision for substitution of additional hydroxyl ions in a non-systematic manner although it has been pointed out by Ross and Hendricks (4, p. 52) that commonly the amount of water liberated above 300° C. is significantly in excess of the amount required by the Hofmann-Endell-Wilm hypothesis.

These generalizations, if valid, indicate that the amount of hydroxyl water of montmorillonite not only exceeds the amount required by the Hofmann-Endell-Wilm hypothesis but the excess amount is related in some way to the distribution of aluminum between octahedral and tetrahedral coordination, as calculated by Ross and Hendricks.

The hydroxyl content can be further considered in light of the thermal decomposition of montmorillonite and beidellite.

ENDOTHERMIC REACTIONS OF MONTMORILLONITE

As a basis for consideration of the structure of this mineral and its relatives, only the liberation of hydroxyl ions is of primary importance. Thus the low temperature endothermic reactions exhibited by montmorillonite (at temperatures below 300° C.) will be largely omitted from the present discussion. Most samples of montmorillonite exhibit two high-temperature endothermic reactions which begin and reach maxima within the approximate ranges of 500° to 750° and 800° to 900° C.

Other interpretations cannot be completely eliminated but, for the purpose of the present hypothesis, these two endothermic reactions are interpreted as activation temperatures required to rupture two different types of hydroxyl bonds within the structural framework of the mineral. Edelman and Favejee (2) have proposed a structure in which hydroxyl ions appear in both tetrahedral and octahedral coordination and this proposal will account for the observed liberation of water in excess of what is required by the Hofmann-Endell-Wilm structure. However, the proposal of Edelman and Favejee will not adequately account for variable amounts of hydroxyl water.

Preliminary experiments, which were designed to examine critically the mode of liberation of water, have led to certain tentative conclusions concerning the thermal decomposition of montmorillonite. Although the experimental results will not be presented here, the conclusions can be briefly summarized as follows:

(1) All of the adsorbed water can be removed at temperatures below 300° C. and, if sufficient time is allowed, at considerably lower temperatures for some montmorillonites.

(2) Approximately the amount of water required by the Hofmann-Endell-Wilm hypothesis is liberated during the temperature interval of the first high-temperature endotherm.

(3) Above the temperature interval of the first high-temperature endotherm, additional water is liberated and the amount seems to be related to the intensity of the second high-temperature endotherm.

Detailed interpretation of these preliminary experiments does not seem justifiable, but it seems probable that the octahedral layer loses hydroxyls during the first high-temperature endothermic reaction and thereby loses some of its structural organization although, judging from the experience of other investigators, its original condition can be partially restored by rehydration (7). During the second high-temperature endotherm hydroxyls are lost from the silica sheets and complete reorganization of the structure is effected during the exotherm which follows almost immediately.

Inasmuch as the occurrence of hydroxyl ions in the tetrahedral layers would require concomitant alteration of the quantities of other constituents within the tetrahedral layers in order to preserve electrostatic neutrality, it has been necessary to find a reasonable mechanism for the introduction of these hydroxyl ions.

THE STRUCTURE OF THE TETRAHEDRAL LAYER

The proposed hypothesis accepts the basic units of the three-layer structure of Hofmann, Endel and Wilm, including the arrangement of the tetrahedra with respect to the octahedral layer; it is concerned essentially with modification of the hexagonal sheet of Si_2O_5 groups, which occasionally contain Al substituting for Si.

In order to reconcile the existing data with a more complete structural picture, it is essential to draw analogies from other compounds which crystallize in aqueous environments at atmospheric temperatures. A particularly useful analogy is the isostructural series:

$Ca_3Al_2(SiO_4)_3$	high temperature
$Ca_3Al_2(SiO_4)_2(OH)_4$	intermediate to high
$Ca_3Al_2(OH)_{12}$	room temperature
$Ca_3Fe_2(OH)_{12}$	room temperature

which has been discussed by Flint, McMurdie and Wells (8), by Pabst (9) and by the writer (10, 11). It has been demonstrated in these studies that $(OH)_4$ has a stable configuration as discrete tetrahedra and 4H becomes equivalent electrostatically to Si in the garnetoid structures. There appears to be no fundamental factor prohibiting this type of tetrahedral configuration in silicate sheets. Extending these considerations to the tetrahedral layers of montmorillonite will permit:

$$n[Si_4O_{10}]^{-4} \rightarrow (n-1)[Si_4O_{10}]^{-4} + [Al_3O_6(OH)_4]^{-7} \qquad (A)$$

or,

$$n[Si_4O_{10}]^{-4} \rightarrow (n-1)[Si_4O_{10}]^{-4} + [Si_3O_6(OH)_4]^{-4}. \qquad (B)$$

These conditions are shown schematically in Fig. 1, where the $(OH)_4$ tetrahedra are shaded. In order to simplify the figure, only the coplanar oxygens with $z=0$ are shown.

It seems desirable to examine some of the principal consequences of this hypothesis:

(1) Hydroxyl ions may occur in the tetrahedral layer of montmorillonite, either substituting simply for Si_2O_5 groups (B) or in association with Al ions substituting for Si ions (A).

(2) Some of the apex hydroxyls of such $(OH)_4$ tetrahedra may be shared with the cations in octahedral coordination and this type of sharing would, in general, take place in the vicinity of Al→Mg.

(3) Although an increase in hydroxyl ions in the tetrahedral layer may

accompany Si→Al, this increase need not be a systematic increase because hydroxyls might enter this layer by mechanism (B).

(4) The sum of the Si+Al ions of the tetrahedral layers need not be constant, but $X+Y+Z/4=4$, if ions of the tetrahedral layers are represented by $(Si_XAl_YH_Z)O_{10}$.

In the description of the theoretical model, an attempt has been made to indicate the isomorphous substitutions in an explicit manner and to examine the precise consequences. Actually one would expect statistical substitutions where real crystals of montmorillonite are involved. Indeed, no greater regularity would be expected for the types of substitutions suggested here than would be expected for such substitutions as Al→Mg in the octahedral layer. Therefore, the association of one $(OH)_4$ group with three atomic replacements of the type Si→Al (Fig. 1) is an arbitrary device which is used merely for the purpose of clearly depicting a general situation. The more general thesis, then, is intended to cover even such statistical occurrences as O→(OH) within the tetrahedral layer; it is not restricted to the intimate association of four such hydroxyls. Nevertheless, in the present hypothesis it is fundamental that the ratio of hydroxyls to silicons in these layers is not constant and that the tetrahedral layer does not have the cristobalite arrangement as postulated by Edelman and Favejee (2).

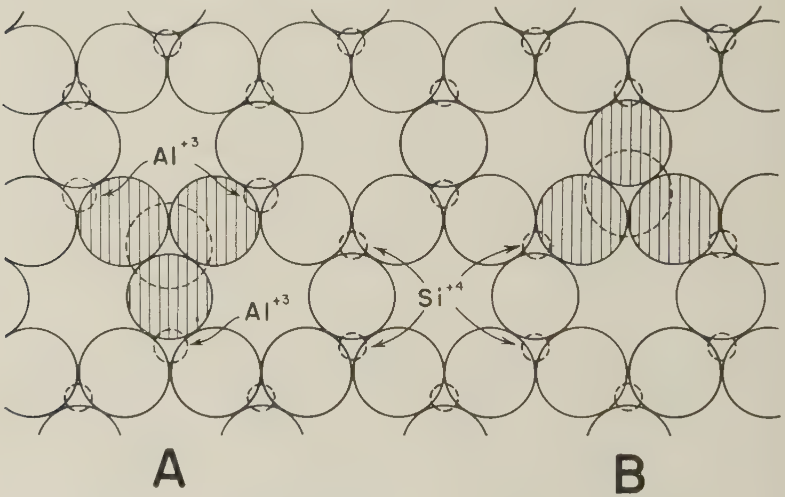

FIG. 1. Arrangement of tetrahedra in the basal plane of montmorillonite, showing the substitutions: (A) $[Si_4O_{10}]\rightarrow[Al_3H_4O_{10}]$ and (B) $[SiO_4]\rightarrow[H_4O_4]$. (NOTE: Only the oxygens with coordinates $z=0$ are shown. Hydroxyls with $z=0$ are shaded. The large dotted circles represent hydroxyls approximately 2.2 Å from the plane $z=0$.)

The general expression for the proposed isomorphous substitutions can be written:

$$n[\mathrm{Si_4O_{10}}]^{-4} \rightarrow (n - m)[\mathrm{Si_4O_{10}}]^{-4} + m[\mathrm{Si}_X\mathrm{Al}_Y\mathrm{H}_Z\mathrm{O}_{10}]^{-R} \qquad \text{(C)}$$

where $X+Y+Z/4=4$, and $R=X+2Y+Z/4=4+Y$. This expression is written in such a way as to preserve the original charge of the silica sheet except for the increase in negative charge which will accompany each Si→Al. Actually these negative charges may be partially compensated by addition of hydrogen ions, and thus it becomes essential to consider the ions of the octahedral layer, as well as the exchangeable bases, in order to obtain neutrality of charges.

THE CALCULATION OF STRUCTURAL FORMULAS

Ross and Hendricks (4) have made a contribution of great value and some of the assumptions applied to their calculations of mineral formulas are undoubtedly justifiable. However, their assumption of 10 oxygens and 2 hydroxyls per structural unit is incompatible with the present hypothesis. For the purpose of testing this hypothesis, it will be necessary to determine quantitatively the hydroxyl water which is associated with different types of bonding energies in order to permit allocation of these hydroxyls to appropriate positions within the structure. Likewise, the amount of Al in tetrahedral coordination will not be calculable until the amount of hydroxyl in the tetrahedral layers has been determined.

In addition to the usual analysis for non-volatile oxides and the precision determinations of water at appropriate temperatures, it will be necessary to determine the base-exchange capacity and the kinds and amounts of exchangeable bases. Although the base-exchange capacity may increase (beyond what is required by Al→Mg) with substitution of Al for Si in the tetrahedral layers, no satisfactory method exists for calculating the inverse relationship, i.e., the amount of this substitution from the base-exchange capacity.

In the present preliminary discussion, the correct procedures for accomplishing these calculations will not be considered. Indeed, it is highly questionable whether sufficient reliable data are available to permit unambiguous calculations of this sort.

CONCLUSIONS

It seems highly probable that the uncertainties and complexities of the montmorillonite structure will be resolved but, in order to accomplish this, analytical data of excellent quality will be required. Analytical data obtained on mixtures, e.g., containing feldspar or quartz, will be quite useless for this purpose and the results will continue to be misleading.

The hypothesis that hydroxyl ions can occur in the tetrahedral layers as substitutions for silicon-oxygen groups has been outlined in general terms. Some of the data which seem to require this modification of the Hofmann-Endell-Wilm structure are discussed, particularly with regard to base-exchange capacity and thermal behavior of these minerals. By this proposal the variable excess of high-temperature water above what is required by the Hofmann-Endell-Wilm hypothesis can be taken into account.

Preliminary thermochemical data tend to substantiate the hypothesis and permit interpretation of the second high-temperature endotherm of montmorillonite as an indication of the presence of hydroxyls in the silica sheets. It is suggested that these hydroxyls can occur in a manner analogous to their occurrence in garnetoids.

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CORDIERITE IN PEGMATITE NEAR MICANITE, COLORADO

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Abstract

The Climax pegmatite of the Micanite district in Colorado contains unusually large masses of cordierite, which is altered in varying degree to pinite, consisting chiefly of muscovite, chlorite, and biotite, and traces of garnet, zoisite, and tourmaline. Data on 50 other occurrences of pegmatitic and vein cordierite are summarized, and the paragenesis of pegmatitic cordierite is discussed.

Introduction

The Micanite pegmatite district lies across the Park-Fremont County line in south-central Colorado, a few miles northwest of the site of the former village of Micanite (Fig. 1). The deposits were investigated briefly by Sterrett (1923, p. 53), who recognized cordierite in the Climax deposit. The pegmatite was studied by the writer in 1946 and 1949, at which times some fresh and much altered cordierite was obtained. This collection was supplemented by material previously secured by Professor K. K. Landes, Chairman of the Department of Geology, University of Michigan. The cost of several thin sections was met by the Department of Mineralogy of the University of Michigan. Thanks also are due to Mr. Marion V. Denny for the photograph of Fig. 2 and to Professor C. B. Slawson of the Department of Mineralogy for a critical reading of the manuscript.

General Geology

The Micanite area is underlain by pre-Cambrian rocks that include granitic gneiss, muscovite gneiss, sillimanite gneiss, mica schist, and biotite granite. Intrusive into the granite and the metamorphic layers are numerous masses of granitic pegmatite, which, in general, are very irregular in shape and structure, with numerous and large apophyses. Zonal structure is developed in some, with conspicuous central units of massive white quartz or of coarse microcline crystals together with massive quartz pods. Concentrations of muscovite occur locally, and it was for this mineral that the deposits first were mined about 1904. In 1934 they were reopened by the Colorado Feldspar Company of Canon City, Colorado, and moderate quantities of good quality feldspar and small amounts of grinding quality muscovite were obtained. Within the district ten major quarries and prospects are recorded (Fig. 1).

The chief minerals of the pegmatites are: quartz, pink microcline in crystals as much as 10 feet on edge, white albite, muscovite in books 2 feet or less in length, biotite, garnet, apatite, magnetite, beryl, sillimanite, cordierite, pinite, hematite (as inclusions in muscovite), bismutite, and an unidentified iron-manganese phosphate.

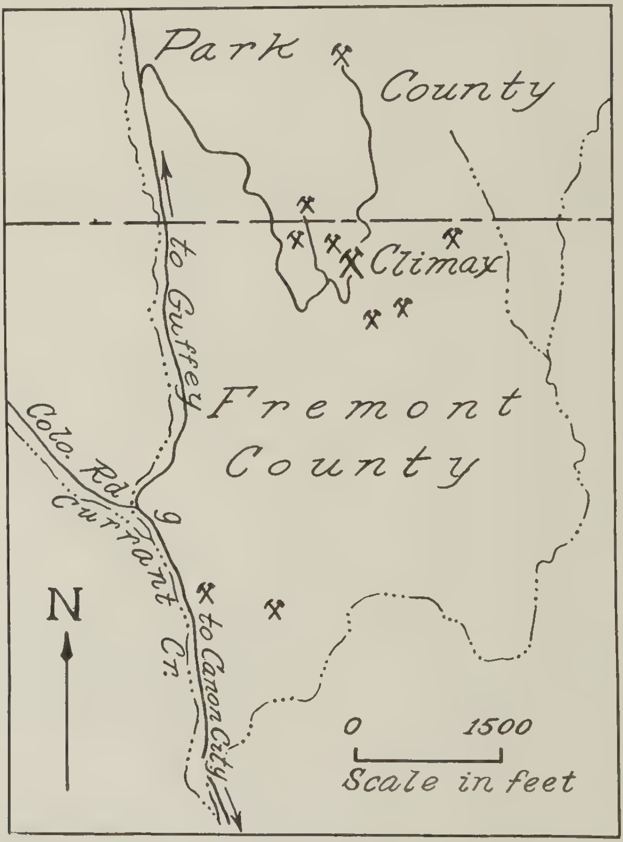


FIG. 1. Index map of part of Park and Fremont Counties, Colorado, showing location of the Micanite pegmatite district and the Climax deposit.

CLIMAX PEGMATITE

The Climax pegmatite is a dike of irregular shape with several large offshoots, which lies in sec. 5, T. 16 S., R. 72 W. on the west side of Micanite Ridge. The body dips moderately to the southeast and trends northeast across the foliation of biotite gneiss, which strikes northwest and dips 40–70° SE. One large apophyse trends north from the northwestern end of the dike, and another extends eastward from the eastern margin. The southwestern end of the dike "horsetails" into several small stringers. Other minor lenses of pegmatite continue the trend of the zone to the southwest. Pegmatite is exposed over a length of 460 feet and a width of 300 feet. The workings consist of eight irregular cuts and pits and a caved adit southwest of and below the cuts, which is reported to be 50 feet long.

The central part of the dike is a large body of massive white quartz whose margins contain veins and radial clusters of muscovite and a few pale green apatite crystals, several inches long. Scattered crystals of microcline as large as 3×4 feet also appear locally along the margin of the core. The quartz is chiefly milky with small patches of clear material. Much of the light brown mica shows "A"-structure and contains scattered to abundant hematite inclusions. Most of the books occur in tabular, moderately-dipping, fracture-controlled replacement masses, one foot or less thick, in which the blades stand normal to the fracture surface. Individual books 1×1½ feet were observed. The chief concentrations of muscovite appear to have been found along the hanging-wall margin of the core. The outer parts of the dike consist of graphically intergrown quartz and dark red microcline cut by large thin blades of muscovite-biotite intergrowth. Specimens of an altered iron-manganese phosphate were found on the dump.

CORDIERITE

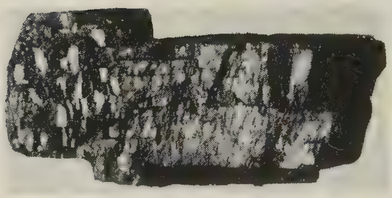
Masses of altered cordierite as much as three feet across are exposed in the wall of the irregular underhand stope from the largest open cut in the southeast part of the dike. They occur in intermediate zone pegmatite along the hanging-wall side of the core and are associated with mica clusters.

The optical properties of the cordierite are:

Optically (–), $2V=50°$, $r<v$, distinct
$\alpha=1.552$
$\beta=1.560$
$\gamma=1.566$

According to the graphs prepared by Folinsbee (1941) these data indicate a cordierite with 60% $Fe_2Al_4Si_5O_{18}$ and 1.5% alkalies.

Most of the somewhat rounded altered cordierite blocks consist of a coarse- to fine-grained mixture of green micaceous minerals commonly referred to as pinite. Relicts of gray to dark violet, glassy cordierite also can be recognized and crystals of red-brown garnet, ¾ inch in diameter are not uncommon. The aggregate is transected by numerous, generally parallel fractures to which most of the micaceous flakes are normal and which represent the *c* {001} parting planes that develop in cordierite with the onset of alteration (Fig. 2). Directly on the fracture surfaces are thin coatings of micaceous flakes lying parallel with the cracks. Irregularly veins of coarse, comb-structure muscovite, $\frac{3}{4}$ inch wide, cut the pinite at various angles.



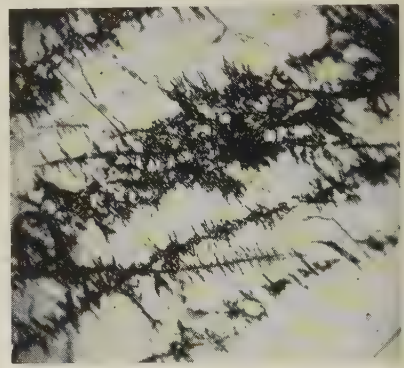
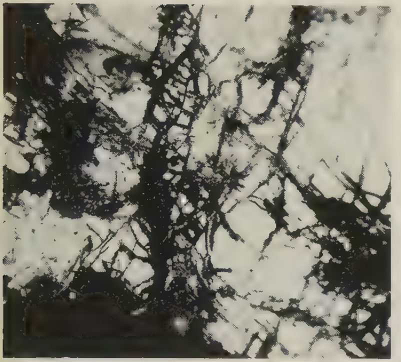


FIG. 2. Specimen of pinite after cordierite, showing development of micaceous blades normal to the {001} parting of the cordierite. Length of specimen is seven inches. Climax pegmatite, Colo.

FIG. 3. Cordierite (light-colored) veined and replaced by pinite. Note parallel stringers along {001} parting. Magnification 20×. Climax pegmatite, Colo.

Microscopically, several stages of alteration can be recognized. The large crystals of cordierite are first traversed by thin, hair-like veinlets of fine-grained muscovite, which send off regular, parallel branches along the *c* {001} parting direction (Fig. 3). In the same stage small radiating clusters of colorless to pale green muscovite flakes develop in widely scattered parts of the cordierite. In the second stage (Fig. 4), subangular pieces of cordierite appear as a replacement breccia, veined and corroded by the micaceous minerals, muscovite, biotite, and chlorite, many flakes of which are stained by limonite. Muscovite is the most abundant of the three, with pale green chlorite next, and a rare flake of biotite, which shows a pleochroism from dark olive green to pale buff. The cordierite relicts are crowded with minute dusty inclusions whose development appears to indicate incipient alteration. In the final stage (Fig. 5) the material consists entirely of a green, shreddy to felted aggregate of fine-grained, plumose muscovite and chlorite with scattered groups of larger muscovite and biotite plates. Here and there occur groups of small, corroded zoisite anhedrons which recall the occurrence of clinozoisite in the pinite of Ebersdorf, Austria (Hlawatsch, 1911). In the final stage tourmaline, pleochroic from pale rose to deep blue, appears in small euhedral crystals (Fig. 6).



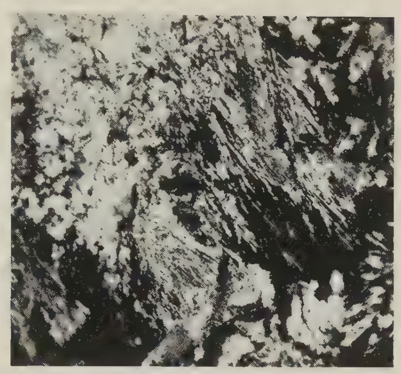


FIG. 4. Angular relicts of cordierite (light-colored) cut by ramifying veinlets of pinite and limonite. Magnification 20×. Climax pegmatite, Colo.

FIG. 5. Pinite, chiefly muscovite and chlorite with minor biotite, pseudomorphous after cordierite, in plumose and dendritic arrangement. Crossed nicols, magnification 20×, Climax pegmatite, Colo.

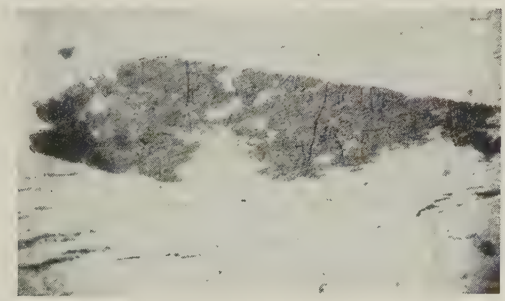


FIG. 6. Tourmaline in pinite after cordierite. Magnification 20×, Climax pegmatite, Colo.

OTHER CORDIERITE PEGMATITES

North America

Cordierite has been found in several other North American pegmatites. Fettke (1914) records it from pegmatites related to the Thomaston granite in southeastern New York. Among the 25 minerals reported from these pegmatites are the aluminous ones, chrysoberyl, dumortierite, kyanite, and cordierite partly altered to pinite. The country rock is the micaceous Manhattan schist. Cordierite from the Haddam, Connecticut, locality has been described by Shepard (1841) and Hovey (1888). The pegmatite, which occurs in biotite gneiss containing cordierite and sillimanite, consists of quartz, microcline, albite, schorl, garnet, zircon, columbite, bismutite, chrysoberyl, and dark blue to purple cordierite, in part pinite. Crystals of gem quality cordierite 5 inches across have been obtained. Cordierite-pinite also occurs in the Collins Hill pegmatite at Middletown, Connecticut (Rice and Foye, 1927) and in the J. Neal pegmatite near Unity, New Hampshire (Shepard, 1841).

Other cordierite-bearing pegmatites from Connecticut have been described by Foye (1922), who records pinite from the Strickland pegmatite at Collins Hill, Portland and cordierite itself in the Bolton schist a few centimeters from the pegmatite contact. The Tims Hill deposit, which contains oligoclase, biotite, tourmaline, allanite, gahnite, and cordierite (altered in part to "fahlunite"), is at the contact of the Haddam gneiss and the Middletown series.

An occurrence at Guilford, Connecticut, usually referred to as a gneiss, consists of blocky masses of purple cordierite, as much as 2½ inches on edge, in a fine-grained, light colored quartz-feldspar-biotite rock.

A dark colored pegmatite in serpentinized peridotite at Winchester, California (Murdoch, 1936), contains oligoclase-andesine, orthoclase, quartz, schorl, biotite, andalusite partly replaced by sillimanite, and bright blue cordierite grains. Pegmatitic lenses in garnet-sillimanite gneiss that occur north of Great Slave Lake, Northwest Territory, Canada (Folinsbee, 1940) contain 4×2×2-inch pieces of gem cordierite, pleochroic from blue to yellow. Ball (1923) notes pegmatites on Upernivik and Langö Islands, latitude 72°47′, Greenland, in which quartz, potash feldspar, garnet, biotite, pyrite, yellow kyanite, and gray-violet cordierite in pieces as large as four centimeters, are the constituents.

Europe

Several cordierite pegmatites are known in Finland. The Nyäng pegmatite in gabbro-amphibolite on Kimito, which has been studied by Eskola (1914, p. 38) and Pehrman (1945) contains the usual granitic pegmatite suite plus spinel, sillimanite, and cordierite, largely pinite (gigantolite). According to Eskola (1914), cordierite also is found in pegmatites associated with a microcline granite at Puurolampi near Orijärvi and intrusive into leptite, granite, amphibolite, and gabbro. The cordierite crystals, altered to pinite, display the faces, (010), (110), (130), and (001). A pegmatite at Degerö near Helsingfors consists of microcline, quartz, oligoclase-andesine, biotite, and cordierite in four-centimeter crystals, partly replaced by pinite (Pehrman, 1932). The presence of previously reported cordierite relicts in gigantolite pseudomorphs of the Tammela pegmatites is denied by Mäkinen (1913), who claims the original mineral was tourmaline. Among the 24 minerals in these bodies are andalusite, chrysoberyl, and gigantolite (cordierite?).

Pehrman (1936) has also discovered cordierite pegmatite in plagioclase-biotite gneiss near Åbo. The minerals are microcline, oligoclase, quartz, biotite, apatite, pyrite, zircon, and cordierite, as much as 3.5 cm. across, bordered by sillimanite needles.

Andersen (1931) notes two Norwegian examples of cordierite pegmatites in his group of pegmatites "endemic in metamorphic rocks." One, on the island of Söndeledfjord, contains oligoclase, quartz, biotite, apatite, hematite, anthophyllite, and cordierite and occurs as lenses in anthophyllite-cordierite-plagioclase gneiss. Another, on a small island west of the island of Hannöen, is in quartz-biotite-garnet schist and consists of quartz, plagioclase, garnet, biotite, apatite, and cordierite.

Kuznetzov (1923, 1930) has described a pegmatite near Lake Uvildy in the southern part of the Kyshtym district, Ural Mountains, U.S.S.R., that contains the accessory minerals, black, blue, and pink tourmaline, garnet, beryl, blue dumortierite, and cordierite. The country rocks are mafic and ultramafic in composition. The cordierite pegmatite of Masslyanka from this area has been classed as migmatic by Fersman (1925). Pegmatites in the Altaï Range near the Caucasian Sea contain cordierite and andalusite with quartz, microcline, muscovite, tourmaline, monazite, rutile, pyrite, molybdenite, powellite, beryl, euxenite, and sericite (Kryzanovskii and Labuntzov, 1926).

In Czechoslovakia cordierite has been recorded from a number of pegmatites. At Velká Skála near Pisek it occurs with albite, apatite, and beryl and is partly replaced by pinite (gigantolite) (Ježek and Krejčí, 1919). Similarly altered, it is found in pegmatite at Wasserhäuseln, Petschau in Bohemia (Gareiss, 1901). Pegmatite at Dolni Bory, Moravia, has the unusual accessory assemblage of tourmaline, chlorapatite, monazite, pyrophyllite, diaspore, andalusite, and cordierite. Both tourmaline and cordierite are partly changed to pinite (Sekanina, 1928, and Jaroš, 1936). Zoubek (1927) has noted a pegmatite in cordierite gneiss at Pelhrimova, with accessory garnet, dumortierite, and cordierite.

In Austria cordierite-bearing pegmatite occurs at Ebersdorf near Pöchlarn in biotite gneiss. The pegmatite minerals are orthoclase, quartz, oligoclase-andesine, muscovite, biotite, schorl, pyrite, dumortierite, andalusite, sillimanite, and cordierite (Hlawatsch, 1911). Pinite, which partly replaces cordierite, contains clinozoisite. Quartzose pegmatitic lenses at two localities in the Selrain Alps, at Montavon and in the Pitzthal, contain plagioclase, muscovite, blue tourmaline, ilmenite, andalusite, and cordierite. Pinite after cordierite contains minor quantities of quartz, chlorite, limonite, epidote, garnet, and kyanite (Gemböck, 1898). The surrounding rock is a biotite schist. A well-zoned pegmatite at Vilshofen on the Danube in Bavaria, Germany, has a core of massive quartz, throughout which are scattered hundreds of pinite "crystals", 10×5 centimeters in size. Although no cordierite relicts were detected (Müllbauer, 1930), recognition of the faces (001), (110), (010), (130), and (100) shows that cordierite was the original mineral. The wall rock is biotite gneiss. A well defined group of aluminous, quartz-rich pegmatites has been recognized in Bavaria by Scholz (1925). One variety, represented by the occurrences at Taferlhöhe by Oberfrauenau, Bärenloch on the Arber, Ansdorf by Hohenwarth, Lengau and Herzogau by Waldmünchen, Düllenberg by Waldassen, and Wintersberg and Katherinenberg in the Fichtelgebirge, contains gigantic individuals of andalusite and much cordierite-pinite as well as biotite, tourmaline, ilmenite, and "uranium mica". Other varieties have sillimanite, kyanite, staurolite, dumortierite, and lazulite.

From Ariège in the Pyrenees, Lacroix (1892) has described a pegmatite with accessory garnet, tourmaline, corundum, and andalusite. An associated quartz vein, also in mica schist, contains tourmaline, andalusite, and large crystals of cordierite-pinite.

Africa

Lacroix (1939) examined at Ankaditany in South Madagascar a pegmatite injected into a quartz-free, biotite-sillimanite gneiss containing local garnet and cordierite. The pegmatite consists of microcline, quartz, almandite, and cordierite in about equal amounts, although in some parts cordierite predominates and biotite is subordinate.

Asia

Near Pipra, in Rewa, India, in a pyroxene granulite consisting of enstatite, diopside, hornblende, biotite, quartz, and plagioclase, Dunn (1929) discovered coarse "segregations" of enstatite-cordierite rock, with minor garnet, sillimanite, quartz, magnetite, and biotite. Brown and Judd (1896) have described a cataclastic, granitic pegmatite from Burma with accessory sillimanite and cordierite.

Shibata (1936) has investigated several cordierite pegmatites in Japan. About two kilometers north of Sasago on the Chûô Railroad is exposed a body of pegmatite containing a fine-grained plagioclase border zone, a wall zone marked by coarse-grained platy biotite, and a core of graphic granite surrounded by a core-margin unit containing andalusite and a graphic intergrowth of quartz and cordierite. This intergrowth is considered to have formed before the crystallization of the graphic granite of the core. Minute dumbell-shaped zircon crystals are included in the cordierite, which is in part altered to pinite. The country rock is a hornfels.

At Higashidani on the west side of Mount Torinomune a pegmatite in amphibolite and cordierite-anthophyllite-biotite schist has an outer zone of oligoclase-andesine, quartz, microcline, and biotite, and a central zone of microcline perthite, quartz, graphic granite, biotite, and cordierite (1×5 cm.). A third pegmatite at Mujinazawa is pipe-like in shape and has been injected into quartz diorite. The outer zone contains plagioclase, potash feldspar, and platy biotite. In the graphic granite core are rosettes of andalusite, schorl, and vugs containing kaolinite, quartz crystals, garnet, pyrite, and sericite. Around the core a graphic granite zone contains biotite and graphic quartz-cordierite rock with included zircon crystals.

Other Japanese occurrences of cordierite in veins or pegmatites are at Hitachi, where the mineral is found in a quartz vein in a copper ore (Kôzu and Watanabe, cited by Shibata, 1936, p. 225); at Naegi in a vein with topaz (same reference); and at Yagen-Yama in Mino Province, where it appears in the margins of banded "granite strings" (pegmatites?) associated with protolithionite, corundum, andalusite, and hercynite (Shibata, 1939).

Australasia

Simpson (1934) has recorded cordierite, partly altered to gigantolite and associated with spessartite in a pegmatite at Melville, Western Australia. Granite pegmatites, cordierite pegmatites, and andalusite pegmatites in peridotite are described by de Jong (1923) from the Kaiboba region on Ceram in the Dutch East Indies.

Discussion

Of the approximately fifty occurrences of pegmatitic or vein cordierite listed in the above summary the type of country rock is not known for 18, but for the rest the wall rock shows considerable variation. Types of biotite gneiss and schist are the most common (10 examples), but hornblende gneiss and amphibolite and their unmetamorphosed igneous equivalents also are represented (6 examples). At only four occurrences is cordierite recorded as being present in the wall rock (cordierite-bearing biotite gneiss and cordierite-anthophyllite rock). Other country rock types include granite, quartz diorite, leptite, granulite, and hornfels.

At a little more than half of the occurrences other aluminous minerals appear with cordierite: andalusite (20 examples), sillimanite (6), dumortierite (4), kyanite, chrysoberyl, and spinel (3 each), corundum (2), and topaz, pyrophyllite and diaspore (one each). Mineral distribution in several deposits shows that pegmatitic cordierite is not one of the earliest minerals to form. It appears to favor cores and core-margin zones, both of which are regarded as having developed at relatively late pegmatitic stages (Heinrich, 1948). Pegmatitic characteristics of the cordierite include coarse grain size, well shaped crystals, and graphic intergrowths with quartz.

The distribution of the occurrences reveals several possible aluminous pegmatite provinces that are characterized by accessory cordierite: (1) southern New England and adjoining New York, (2) southwest Finland, (3) Czechoslovakia and Austria, (4) Bavaria, and (5) Japan. About a dozen other localities are scattered throughout the world.

Theories seeking to explain the origin of pegmatitic cordierite may be listed as follows:

1. *Endogenic origin*
    - (a) By inclusion and some recrystallization of material from adjacent, cordierite-bearing wall rocks. Such cordierite is xenocrystic.
    - (b) By the incorporation of aluminous country rock at depth by granitic pegmatite magma. Such cordierite is of pyrogenic formation in a contaminated magma.
    - (c) By the interaction of quartz-diorite pegmatite magma with amphibolite (Shibata 1936).
2. *Metamorphic origin.* By metamorphic differentiation, i.e., migration, concentration, and recrystallization of mineral substance originally disseminated in wall rock.
3. *Magmatic origin.* By direct crystallization from a pegmatitic magma characterized by a high primary content of aluminum. Such cordierite is of pyrogenic origin from an uncontaminated magma.

The first hypothesis, that of inclusion of wall rock material, has little to recommend it. Apparently cordierite is not a common constituent of the wall rocks of cordierite pegmatites. Moreover the mineral apparently prefers to form in the interior rather than the exterior parts of pegmatites. The mingling of pegmatitic magma with aluminous rock at depth has been a convenient means of explaining an excess of aluminum and can neither be proved or disproved in most cases. The regional distribution of accessory cordierite in some pegmatites may be a point against this hypothesis. One might also expect to find some definite evidence of contamination, possibly relicts of reworked inclusions or a considerable variation in the chemistry of certain pegmatites in a district. Fersman (1931) includes cordierite pegmatites in his Group IV, contact pegmatites, which he regards as contaminated pegmatites. Shibata (1936) ascribes the cordierite in pegmatite north of Sasago to this origin but later states that the mineral at Higashidani was formed by the interaction of quartz diorite pegmatite and amphibolite. The cordierite at Mujinazawa also is explained by this latter mechanism, even though no amphibolite exposures are recorded there. Such an interaction of basic country rock and intermediate magma is not likely to produce a pegmatite of granitic composition with a slight excess of aluminum and magnesium.

An origin by metamorphic differentiation is implied by Andersen (1931), who includes cordierite pegmatites in his group of pegmatites "endemic in metamorphic rocks". A similar origin is suggested by Dunn (1929) in his use of "segregation" for the occurrence at Pipra, Rewa, India. Metamorphic differentiation as outlined in the literature has always been but a vaguely defined term. The process, as sketched, appears to be analogous to a high intensity type of lateral secretion, and is believed to include selective subtraction of certain constituents from a large volume of rock, their migration to certain favored loci, and recrystallization as considerably larger mineral grains. Evidence for operation of these mechanisms is lacking in the case of cordierite pegmatites.

Cordierite is listed as an accessory pegmatite mineral by Ossan (1923) without mention of the possible mode of its formation. Ford (1932, p. 583) states, "Cordierite may be the direct product of igneous action, being stable over a considerable range of temperature and formed apparently without necessarily the aid of mineralizers." Johannsen (1932, Vol. II, pp. 166–167) writes, "Among the granites it (cordierite) is confined almost exclusively to the two-mica varieties. In some cases it is certainly primary; in others it is clearly metamorphic". Factors favoring the theory of pyrogenic formation of cordierite from an uncontaminated magma are: the occurrence of many cordierite pegmatites in provinces, the position of the mineral within pegmatites, its typical pegmatitic characteristics, and the absence of evidence of assimilation in most cordierite pegmatites.

Conclusions

(1) Cordierite occurs in granitic pegmatites of two types: those containing other aluminous minerals, chiefly andalusite and sillimanite, and those in which cordierite is the only aluminous accessory.

(2) There appears to be a tendency for cordierite pegmatites to occur in provinces.

(3) The country rock around cordierite pegmatites usually does not contain cordierite, although lack of detailed data on country rock mineralogy may affect this conclusion. Wall rocks vary from acid to basic and are both igneous and metamorphic.

(4) The theory of crystallization from an uncontaminated pegmatite magma has favorable supporting arguments, but crystallization from a magma contaminated through assimilation of aluminous rock at depth cannot be dismissed in all cases. Cordierite that occurs near pegmatite margins may represent xenocrystic material.

(5) Cordierite that crystallized in the magmatic pegmatite stage is not in equilibrium during the hydrothermal stage and is altered to pinite, which consists chiefly of muscovite, chlorite, and biotite in varying pro-

portions. This change requires chiefly the introduction of potassium and hydroxyl by pegmatitic solutions.

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DISORDER IN THE MINERAL BORNITE, Cu_5FeS_4

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Abstract

X-ray data from a single crystal of Bristol, Connecticut, bornite shows that bornite is a pseudocubic orthorhombic crystal with the following cell dimensions.

$$a=21.94\ \text{Å}$$
$$b=21.94\ \text{Å}$$
$$c=10.97\ \text{Å}$$

X-ray, thermal, and electrical evidences indicate that bornite can exist in both a low- and a high-temperature form, and that the structural difference between the forms is one of order-disorder. When thermally induced, the disorder first reaches a measurable magnitude at 170° C. and the critical temperature of complete disorder is reached at 220° C. By quenching in cold water the high or disordered form can be retained, but if cooled slowly from the elevated temperature the low form is regained.

The high-temperature form is cubic with a cell edge of 10.97 Å and belongs to one of the space groups: $F\bar{4}3m$, $F43$, $Fm3m$.

Introduction

At a recent meeting of the Crystallographic Society of America the author presented a paper on "Disorder in Bornite, Cu_5FeS_4." Shortly thereafter it was called to his attention by Professor George Tunell that there was a paper already in press on "The Symmetry and Crystal System of Bornite" by G. Tunell and C. E. Adams [1] in which some of the data seemed to be in disagreement with that reported by the author. Because of this disparity it seemed advisable to present a more detailed account of the study of disorder in this mineral at the present time.

Historical Background

Bornite usually occurs as a hypogene mineral intimately associated with chalcocite (Cu_2S) and chalcopyrite ($CuFeS_2$) [2]. Many intergrowths of these minerals as well as intergrowths of tetrahedrite ($Cu_{12}Sb_4S_{13}$) and klaprothite ($Cu_6Bi_4S_9$) in bornite have been described in the literature [3, 4, 5, 6, 7, 8, 9] and the conclusion has invariably been reached that the intergrowth textures found are the result of the unmixing of these different minerals from bornite. In many instances heat-treating experiments were conducted to demonstrate that the exsolved plates could be redissolved in the bornite host at elevated temperatures and that subsequent exsolution would occur under the proper cooling or annealing conditions. G. M. Schwartz [4] showed that the chalcopyrite lamellae in a naturally occurring bornite sample would dissolve into the bornite when the temperature was raised to 475° C. and that by quenching in cold water the solid solution could be retained at room temperature. R. Latsky [6] demonstrated that exsolution occurs in bornite at a lower temperature than that at which dissolving takes place, and that a sample of bornite in which the lamellae of chalcopyrite have been completely dissolved and "quenched in" will, when held at 220° C. for seventy-two hours, exhibit exsolution blebs and minute specks of chalcopyrite. A. B. Edwards [7] was able to redissolve the lath shaped intergrowths of tetrahedrite existing in the bornite from Pine Valley, Australia, at a temperature between 250° and 285° C.

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Thus it has been clearly demonstrated that at elevated temperatures the structure of bornite will accept additional iron and copper atoms, and will permit the diffusion of these atoms through it, and, that at lower temperatures there is a return to stoichiometric proportions by the expelling of any excess atoms in the form of exsolved material. Therefore, it might be expected that self diffusion may also occur in bornite at these elevated temperatures, and, as M. J. Buerger [10] has pointed out, "the process of ordinary self diffusion in a crystal resembles that of substitutional disorder."

A determination of the crystal structure of bornite was attempted but not completed by D. Lundqvist and A. Westgren [11] from powder photographs of synthetically prepared material. They found bornite to be cubic, space group *Fd*3*m*, with a cell edge of 10.93 Å. The structure they proposed is based upon sulfur atoms arranged in cubic close packing (face-centered cubic) with the metal atoms placed in the interstices. Several distributions of metal atoms were suggested but none of them could be definitely confirmed by *x*-ray intensity checks.

More recently G. Tunell and C. E. Adams [1] have obtained rotating crystal and Weissenberg data with a single crystal of bornite from the Carn Brea Mine, Illogan, Cornwall, and found the true cell edge to be 32.81 Å. By adopting a cubic cell of edge length 5.47 Å with the sulfur atoms at the origin and at the face centers (close packed cubic) they found reasonable intensity checks for all of the spots of strong and medium intensity and some of the spots of weak intensity when four copper or iron atoms were statistically distributed over the positions $\frac{1}{4}\frac{1}{4}\frac{1}{4}$, $\frac{3}{4}\frac{3}{4}\frac{1}{4}$, $\frac{1}{4}\frac{3}{4}\frac{3}{4}$, $\frac{3}{4}\frac{1}{4}\frac{3}{4}$, and two copper or iron atoms were statistically distributed over the positions $\frac{1}{4}\frac{3}{4}\frac{1}{4}$, $\frac{3}{4}\frac{1}{4}\frac{1}{4}$, $\frac{1}{4}\frac{1}{4}\frac{3}{4}$, $\frac{3}{4}\frac{3}{4}\frac{3}{4}$, the total number of copper and iron atoms averaging five per cell and one per cell respectively. However, a considerable number of spots of weak intensity were not accounted for by this approximate arrangement.

MATERIAL

Material for the present investigation was obtained from the Harvard University mineralogy collection through the courtesy of Professor Clifford Frondel and was selected for *x*-ray, thermal, and electrical investigation only after polished section examination showed the material to be relatively homogeneous and free from exsolved or included extraneous matter. Only one locality, that of Bristol, Connecticut, provided single crystals of useful dimensions and quality; some additional, fairly pure bornite from Butte, Montana, was selected.

X-RAY INVESTIGATION

A complete diffraction record was taken of a single crystal from Bristol, by the rotating-crystal and Weissenberg methods. This record indicates that the Bristol bornite in its low-temperature form is a pseudo-cubic orthorhombic crystal with the following cell dimensions:

$$a=21.94\ \text{Å}$$
$$b=21.94\ \text{Å}$$
$$c=10.97\ \text{Å}$$

The *a* axis and the *c* axis rotation photographs are shown in Figs. 1*a*, and 1*b*, respectively. The rotation photograph of the *a* axis appears to be identical with that of the *b* axis, but a closer study of the upper levels of the *c* axis by the Weissenberg method indicates that the crystal does not possess a four-fold axis, but is based upon a primitive orthogonal cell. Due to the fortuitous lack of reflections on the zero and first levels, the presence or absence of glide planes and screw axes could not be determined with certainty by systematic extinctions.

The identical crystal used in obtaining the record described above was heated in a sealed evacuated pyrex tube to 225° C. and quenched in ice water. The crystal was mounted and oriented parallel to an axis previously identified in the low-temperature form as the *a* axis and a rotating crystal photograph was taken (Fig. 1*c*). From this photograph and the supporting Weissenberg photographs it is evident that through heat treatment the crystal becomes isometric with a cell edge of 10.97 Å. The diffraction symbol is $m3mF$— and hence there are three possible space groups under which the symmetry of the heat treated crystal may be classified: $F\bar{4}3m$; $F43$; and $Fm3m$.

Structurally the difference between the low-temperature and high temperature form of the Bristol bornite lies in the presence of a superstructure in the low form. Supported by the solid solution data in the literature as outlined above, and by the thermal and electrical evidences described below, this would indicate that self diffusion and subsequent

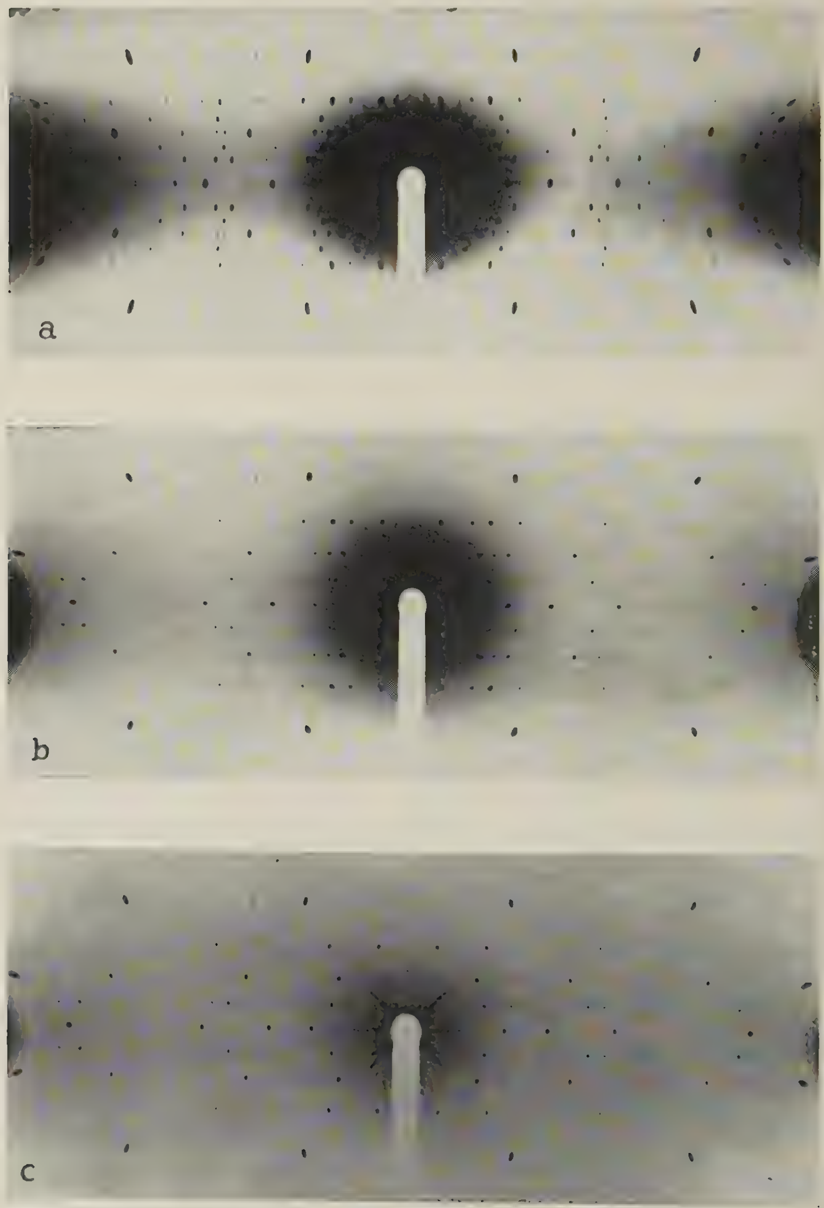

FIG. 1. Rotating crystal photographs of bornite from Bristol, Connecticut, made with cobalt Kα radiation (Iron filter). (*a*) Rotated about the *a* axis. (*b*) Rotated about the *c* axis. (*c*) Identical rotation as (*a*) above only after crystal had been heated to 225° C. and quenched.

substitutional disorder is occurring at the elevated temperature. That the difference between the high and low forms is just one of simple substitutional disorder between the iron and copper atoms is unlikely. If this were the case then all the reflections that disappear as a result of the heat treatment, and which constitute the superstructure reflections, would owe their intensity to the difference in scattering power between iron and copper atoms. It would normally be expected that these reflections would all be very weak, as the difference in scattering factor between iron and copper is only 10% at low Bragg angles increasing to 14% at higher angles. Such strong spots as those that exist on the third and fifth levels of the ordered crystal are difficult to justify in view of this small difference in scattering power. Therefore, either slight shifts of atomic parameters within the cell might also be expected, or vacant sites or interstitial spaces are also involved in the disordering.

Lundqvist and Westgren [11] have already suggested the possibility that the metal atoms are distributed at random throughout the interstices of the close packed sulfur structure. However, as the cell edge of the high-temperature form has been reduced to 10.97 Å, rather than to 5.49 Å the edge length of a single close packed sulfur cell, either a selected number of interstitial sites must be involved or a selected number of atoms are involved while the others remain in definite positions. An example of the latter case would be realized if the iron atoms remained fixed in position while the copper atoms are statistically distributed throughout the remaining interstices.

Thermal Investigation

As crystals in their disordered form are at a higher energy level than when in their ordered form, the change of bornite from ordered to disordered with increasing temperature is accompanied by a gain in energy that is evidenced by the absorption of heat by the crystal from its surroundings. Thus the differential thermal analyzer record for both Bristol and Butte bornite showed a definite endothermic saw-tooth shaped peak that would be expected to accompany disorder (Fig. 2). The deflection of the peak first starts at about 170° C. and is completed by 220° C. At 260° C. a rather broad exothermic peak is produced. From the presence of sulfur dioxide fumes and from the appearance on the x-ray powder record of the product of additional reflections representing new phases, it is evident that the exothermic peak is due to the dissociation and subsequent oxidation of the sulfide. If the sample is rapidly cooled from a little above 220° C. the endothermic peak is not present when the run is repeated. However, if the sample is cooled slowly from 220° C. the run can be repeated with the record being reproduced identically except for a slight loss in the amplitude of the endothermic peak.

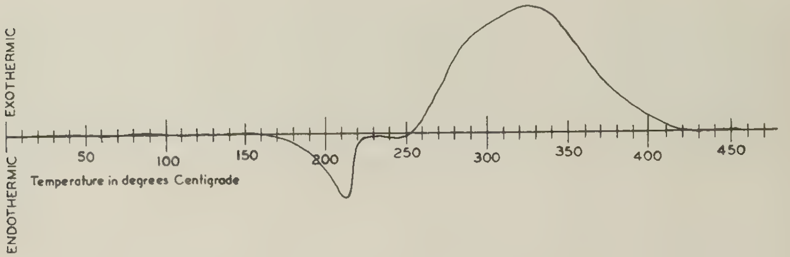

Fig. 2. Thermograph of bornite from Bristol, Connecticut. Recorded during a thermal rise of 12° C. per minute.

Resistivity

Changes in electrical properties can be expected to accompany an order-disorder change [12, 13]. Although marked changes can also be expected in thermoelectric potential, resistivity is the electrical property most amenable to measurement at elevated temperatures. Little blocks of bornite 7 mm. × 1 mm. × 1 mm. were formed on the polishing lap from samples of both Bristol and Butte material. Resistivity measurements were made on these blocks over a range of temperature from 30° to 260° by measuring the voltage drop across a constant section of the block with a potentiometer while the current through the block was maintained at a constant level. A plot of temperature versus resistance (Fig. 3) exhibits a smooth curve of decreasing resistance with increasing temperature until 170° C. is reached, thus indicating that bornite throughout this temperature range behaves as a semi-conductor [14]. At 170° there is an increase in the rate of decrease of the resistance, smoothing out again after 220° C. is reached.

Reversibility

The ability for the disordered compound to reorder upon slow cooling was indicated by the reproduceability of the thermal record after slow cooling as described above. This reversibility has been verified by means of *x*-ray powder photographs. On the complete powder diffraction record, when taken with cobalt radiation, there are only two superstructure lines, 362 and 347, that are of sufficient intensity to show above the background. These two reflections are missing from the diffraction record of bornite after the powder has been quenched from 225° C. in a manner identical to that described for a single crystal. When the bornite powder

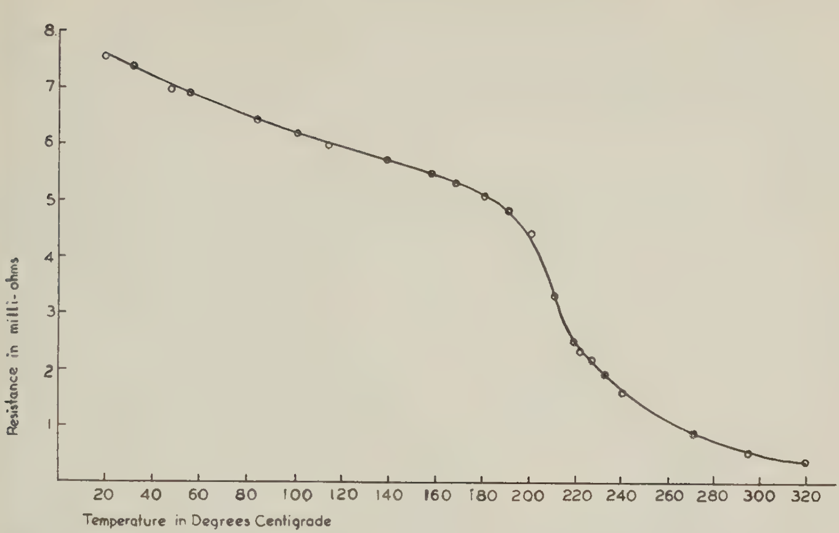

FIG. 3. Resistivity versus temperature of bornite from Butte, Montana. Measurements made across a constant section of material at a constant current of 12 milli-amperes.

in which the disorder has been "quenched in" is reheated to 225° C. and allowed to cool at the rate of 10° C. per hour to room temperature the x-ray powder diffraction record shows the reappearance of the superstructure lines, thus indicating a return to the low temperature form.

SUMMARY

From the results of this investigation it is evident that bornite can exist in at least two structurally different forms, and that the relation between these forms is one of order-disorder of the metal atoms. When thermally induced the disorder reaches observable proportions at 170° C. and the critical point of complete disorder is reached at 220° C. The disorder can be accounted for by one of the following mechanisms:

1. Substitutional disorder between some or all of the metal atoms with slight readjustments of the atomic parameters to account for the changes in the intensities of some of the x-ray reflections.

2. Substitutional disorder between some or all of the metal atoms and some or all of the interstitial openings in the close packed sulfur structure.

ACKNOWLEDGMENTS

The writer wishes to thank Professor Martin J. Buerger for his counsel during the course of this research, and for critically reading this paper. Thanks are also extended to Professor George Tunell for his courtesy in providing the writer with a pre-publication copy of his paper [1].

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TWINNING IN THE DIAMOND

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ABSTRACT

The geometrical concept of twinning in the diamond is inadequate because a reflection across either (111) or (11$\bar{2}$) will give identical relationships. The twinning plane can be identified by observing the contact or by demonstrating structurally that (111) lends itself to the growth phenomena involved in twinning. In either normal or twinned growth across (111) the first and second coordinations are identical and differences appear only in the third or higher coordinations. The statistical opportunity for twinned structures to be initiated is therefore high.

The probability of atoms falling into the normal or twinned position is not wholly a function of the structure. The surface energy distribution on a growing crystal face may be modified by the type of impurity that forms an adsorbed film on the surface. The resultant energy pattern may increase or decrease the statistical probability of the atoms attaching themselves in the normal or twinned position. By slowing the rate of migration of atoms to the surface the impurity film will promote the orderly accretion of atoms. Structural conditions indicate that diamonds do not grow by the accretion of single atoms.

Twinning is highly characteristic and what are apparently random intergrowths are more often multiple twins. The classical examples of contact, penetration, polysynthetic, and cyclic twins, by focussing attention on the geometrical symmetry of highly unusual and non-characteristic growth forms, obscure the true structural basis of twinning. Tables useful in the analysis of complex twins of non-symmetrical external form are given.

Crystallographic continuity exists across a twinning plane. All other boundaries between twinned individuals are suture contacts which are very often the loci of voids and inclusions.

INTRODUCTION

Twinning is commonly defined in terms of a geometrical relationship which exists between the individuals comprising the twin. However, in the cubic system, a geometric concept alone is inadequate because more than one plane of reflection may describe the geometric relationship between two crystals which are twinned with respect to each other. This condition arises because of the equally spaced coordinate system.

If the diamond is assumed to have hexoctahedral symmetry, its twinning may be defined on a geometric basis as (1) a reflection across the rational plane (111) or, as (2) a reflection across the rational plane (11$\bar{2}$). These planes are mutually perpendicular, and both are also perpendicular to a secondary plane of symmetry. Hence, either plane will serve to define the relationship which exists between the twinned individuals (Fig. 1). There are three planes of the type (11$\bar{2}$), and in the idealized interpenetrating cubes (Fig. 2) after the spinel law (which are common in diamonds from the Belgian Congo) we have a total of four planes of symmetry, any one of which may be a twinning plane if such a plane is defined on a geometric basis only. Also, any of the four axes normal to these planes may be the twinning axis. This twinning may also be defined geometrically as a rotation of 70° 32′ about a binary axis of symmetry. This geometrical operation, however, does not meet the accepted concepts of a twinning law.

Twinning is not a geometric phenomenon but a deviation in the normal process of crystal growth. While this deviation gives rise to a discontinuity in the crystal structure, it is a controlled deviation—an ordered process—and therefore we have crystallographic continuity across the twinning plane from one individual to the other. The twinning plane is that plane along which the ordered deviation in the crystal structure takes place. In the diamond it is the (111) plane. In the microscopic sense twinning is typical and characteristic of the diamond. No single diamond that has been subjected to detailed examination has been found free from twinning of the mosaic type.

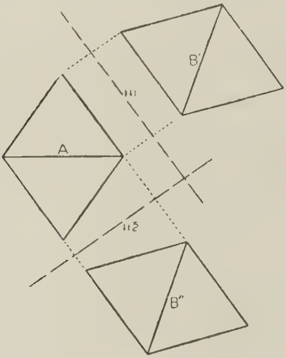


Fig. 1

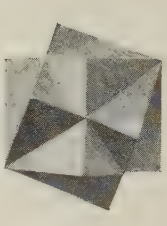


Fig. 2

Much of the confusion in regard to twinning arises from the classical conception of twinning as a geometrical phenomenon. Given the complete goniometric measurements of a twinned diamond, it would be impossible to determine whether the twinning law was (111) or ($11\bar{2}$). The decision as to which plane is the twinning plane can be made by observing the twinning contact between the two individuals. This contact plane may generally be readily observed in contact twins, but in such substances as fluorite, which occurs only in "penetrations twins" (Fig. 2), no positive determination can be made except by observing the contact on an oriented polished surface. This condition arises because the crystal habit of fluorite is cubic while the twinning is octahedral. In doubly refracting crystals the two individuals can be easily differentiated, but in singly refractive substances the presence of twinning other than that revealed by gross macroscopic features often goes unobserved. Figure 3 shows fine suture-like twinning in the diamond which has been revealed by its hardness variation. Twinning contacts in quartz are brought out by etching a polished surface, revealing the "electrical twinning," which is not differentiated by double refraction.

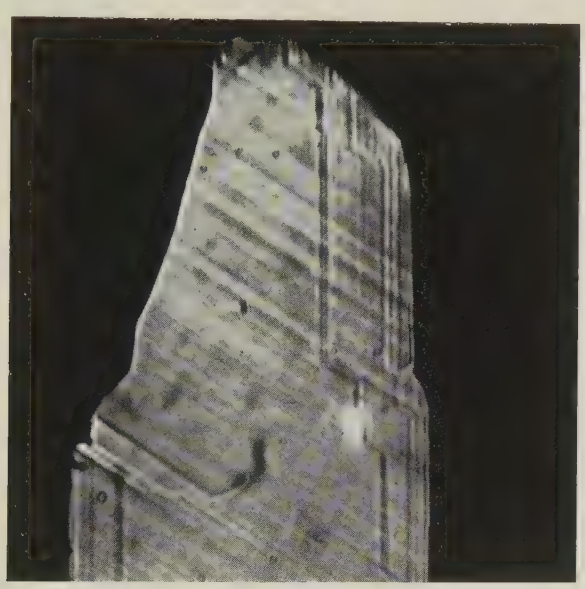


Fig. 3.

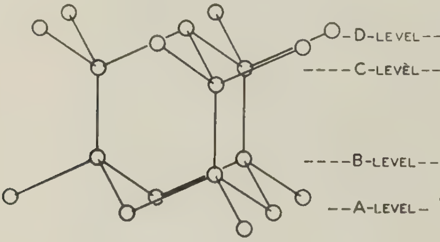
Polishing lines run lower right to upper left. Twinning lines vertical.

Where two geometrically possible twinning planes exist the choice can generally be made from structural considerations. One plane will have an energy distribution such that twinning across this face will be more probable than across the other.

STRUCTURAL TWINNING

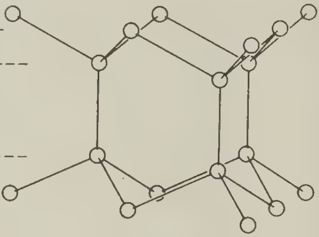
In Figs. 4 and 5, four layers of atoms in the normal and the twinned position are represented with the octahedral face in the horizontal plane. In the twinned structure all primary bonds are completely satisfied and no free energy develops at the interface as a result of the relative positions of the *B* and *C* layers. The crystal is therefore continuous across the interface or twinning plane, although there is a discontinuity in the structure. The twinning plane is parallel to (111) but cannot be definitely localized, because the *B* and *C* levels of atoms may be considered as lying in either the upper or lower individual.

For purposes of the discussion, it is desirable to consider that the twinning plane is located between the *B* and *C* levels. Thus identified, the adjacent octahedral surfaces of both individuals, normal and twinned, consist of carbon atoms arranged in equilateral triangular patterns held together by bonds normal to the surface. If any atom in the C level of the normal structure (Fig. 4) be taken as a center and the C and D levels be rotated sixty degrees, a structure identical to the twinned structure (Fig. 5) would be developed. Each atom of the third layer would still have its counterpart directly above it.



NORMAL

Fig. 4



TWINNED ON (111)

Fig. 5

In the growth of a crystal by accretion on the octahedral surface a discrete aggregate of ordered atoms drifting toward the octahedral surface would have two positions, sixty degrees apart in which it could attach itself to the crystal. The distribution patterns of surface energy on the contacting surfaces determine which of the two positions, normal or twinned, would be chosen. The outermost layer of atoms on each surface are identical and would make identical contributions to the distribution patterns of surface energy. They would be dominant in forming this pattern. The layer of atoms next to the surface would produce a secondary trigonal distortion in the distribution pattern at the surface (Figs. 6 and 7).

Buerger[1] has expressed a somewhat similar idea in stating that the higher coordination spheres control twinning. In the diamond structure it can be demonstrated that both the primary and secondary coordinations are identical in both the normal and the twinned positions, and that variation is introduced only in the tertiary coordinations. Its influence is so remote that it seems most unlikely that it could affect the structure. But in the case of surface energy distribution patterns the layer of atoms immediately below the surface would exercise a considerable influence upon the patterns.

In Figs. 6 and 7 an attempt has been made to represent schematically

[1] *Am. Mineral.*, **30**, 172 (1945)

the trigonal pattern of energy distribution produced by the surface and second layers of atoms on a crystal face. If a crystal grows by the addition of groups of atoms rather than single atoms the primary bonds of the surface layers will be equally as well satisfied by the normal or the twinned position. They will also exercise the dominant control in pulling the two surfaces together in one or the other of the two positions, which are sixty degrees apart. An aggregate of atoms drifting toward a crystal face in the halfway or thirty degree orientation must rotate to one or the other of the bonding positions. The polarity of the surface energy distributions as far as it is determined by the surface layers of atoms will be identical and would exercise no control over the attachment of drifting atoms in either the normal or twinned position. The control must come from the contribution to the surface pattern made by the second layer of atoms, and it might be assumed that if the approach was midway between the bonding positions, the aggregate would rotate to the normal position and attach itself to the crystal. There might be some intermediate position of approach where the drifting aggregate could take the lesser angular rotation toward the twinned position rather than the greater toward the normal bonding. On a structural basis alone one might assume that whether this neutral position is at 15°–45°, 10°–50°, or some other point, would determine the statistical prevalence for the normal or the twinned position.

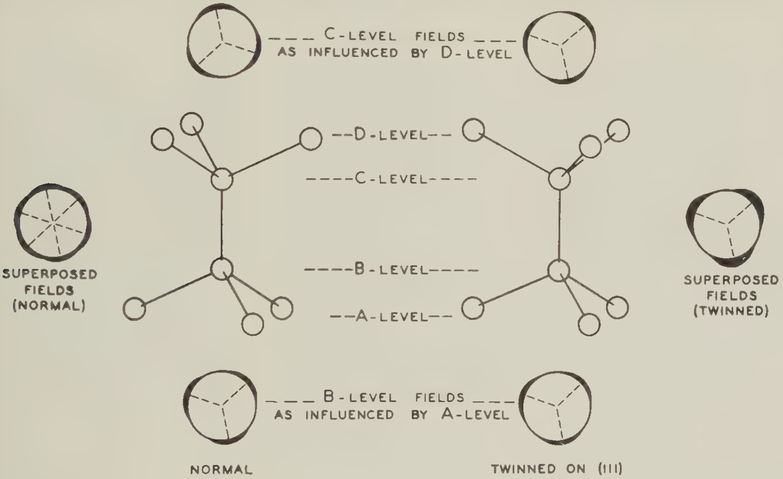


FIG. 6

FIG. 7

Such an argument as that just given assumes that the surface energy distribution of a crystal is a function of its structure only and does not take into consideration the nature of the environment in which the crystal is growing. The unsatisfied bond energy at the surface of the crystal attracts and holds selectively molecules or atoms from the environment in which it exists. This film not only modifies the energy distribution of the surface but serves as a barrier through which the attaching atoms must migrate. A corresponding film on the surface of the arriving atoms exercises a similar control. The character of the film and consequently its influence will vary with the character of the environment. The choice then of whether the arriving atoms will join the nucleus in a normal or twinned position becomes dependent also upon the "reaction" that develops between the crystal face and the environment.

It appears to be necessary to have crystal growth on the octahedral surface of the diamond proceed not by the addition of a single layer of atoms, but at least by a double layer. From Figs. 4 and 5 it can be seen that if the *C* layer were added to the *B* layer, the resulting surface would have three times the free surface energy it had previously (three unsatisfied bonds per surface atom instead of one). By the addition of a double layer of *C* and *D* atoms the free surface energy remains the same. It appears therefore necessary to have crystal growth proceed by the accretion of two or multiples of two layers of atoms. This naturally holds true for both the normal and twinned positions.

Twinning Morphology

Because classical crystallography was founded on the external morphology of crystals, the concepts of contact and penetration twins developed. While the penetration twin may still be useful in describing some of the external features of twinning, and therefore assist in the recognition of minerals which are found characteristically twinned, it has no structural or theoretical significance. The spatial patterns of atoms cannot interpenetrate, and twinned crystals can only be in contact with each other. Interpenetrating twins of diamond are common only in the Belgian Congo production. These crystals are of the type shown in Fig. 2 and are usually yellow to brown in color and opaque. Occasional twins of this type are glassy, colorless to white, and may be as large as one hundred carats or more.

Gross manifestations of twinning represent exceptional cases where the two individuals have grown to approximately the same size. The De Beers Consolidated Mines, Ltd., give the percentages of macles (twins) of all stones recovered from the Dutoitspan, Bultfontein, and Wesselton Mines as 7.39, 5.25, and 3.71 per cent, respectively, or an average of 4.37 per cent. The percentages of "flats" (which are commonly twinned) from these mines are 9.05, 5.63, and 11.21, or an average of

8.63 per cent. Approximately 40% of the production consists of industrial stones which are not sorted into twinned and untwinned classifications. Macle is a classification of gem quality stones from which a fair recovery of polished stones can be made. They may be parted along the twinning plane by a process practically identical with cleaving. It is the impression of cleavers that they "cleave" along the twinning plane slightly easier than along a true cleavage, but that the resulting surfaces are apt to be more irregular. With such a high percentage of diamonds showing gross evidence of twinning, it is not surprising that microscopically all diamonds have internal twinned zones or areas.

FIG. 8

FIG. 9

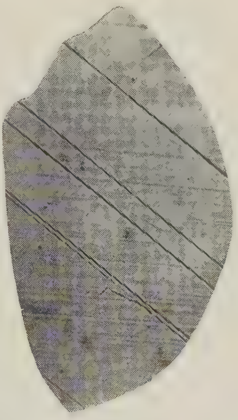


FIG. 10

Polishing lines are nearly horizontal.

Figure 8 shows the polished surface of a diamond which has been lapped in such a direction that the directional variation in hardness brings into relief the twinned zone. This zone itself is further twinned. The orientation of the twin differs from that of the surrounding diamond, and when it is being lapped on a softer direction, the twinned zone presents a hard direction to the lap and resists abrasion. When viewed in the reflecting microscope, the beveled edge along the twinning contact stands out in relief, because it does not reflect the vertically incident light uniformly with the rest of the flat surface. This relief is accentuated when the microscope is slightly out of focus and appears to move parallel to the contact when the tube of the microscope is raised and lowered in a manner similar to the Becke line. When the tube is raised, the line moves towards the area in lower relief, in this case, away from the twinned zone.

The diamond cutting profession has developed a terminology to designate the various types of cross-grained structure (twinning) that causes difficulties in polishing a diamond. When the diamond cutter is lapping on the softer or more rapid cutting direction of the main part of the stone, the twinned area is presenting a hard direction, and vice versa. By choosing an intermediate direction, he finds one in which the two areas both cut at the same rate. If a third area is twinned in a direction not parallel to either of the other areas, it is impossible to remove all evidence of the twinning on a polished surface.

The penetration and contact twin of the mineralogist is called a *macle* by the diamond cutter. He designates as a *block* an enclosed twin that appears as a regularly shaped area on a polished surface (Fig. 9). In the early stages of the growth of the twin one individual has become dominant and completely encloses the other. A *knot* is an enclosed twin of irregular outline and often two or more individuals comprise the knotted area. A *ping* is a small speck-like area that stands out on the polished surface and from which a comet-like tail usually streams in the direction of lapping. It can be demonstrated that all these types are twinned on (111) by the trace of the twinning contact on an oriented polished surface or by measuring the angle between the trace and the lapping direction at which host and the twin abrade at equal rates. Contact twins rarely consist of only two individuals but commonly are polysynthetic, at least in the region where it is macroscopically evident that the twinning plane lies. Microscopically, the commonest types of twinning observed are the speck-like inclusions (*pings*) and paper-thin layers which on a polished surface appear as continuous (Fig. 10) or discontinuous (Fig. 3) lines parallel to traces of the octahedron. A single crystal commonly reveals twinning on more than one of the octahedral faces. Complex twins such as that in Fig. 11, a seventy-five carat diamond consisting of four cubes, are classified in the bort category and

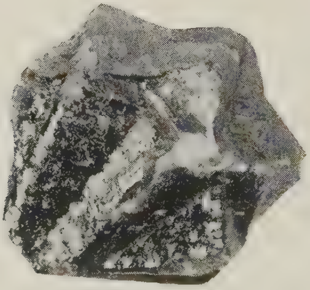


FIG. 11

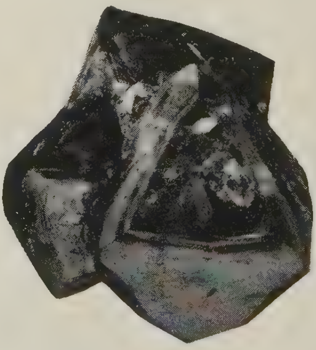


FIG. 12

crushed into grit for grinding wheels. Much of the bort that at first glance appears to be a random intergrowth can be demonstrated to be complexly twinned.

Most of the literature on twinning deals with crystals of the lower symmetry classes. In the case of the plagioclase feldspars, repeated twinning on (010) gives rise to polysynthetic twins with a distinctive geometric pattern, because there is only one direction of growth which the twinned crystal may take. However, in rutile, which twins on (101), there are four structurally equivalent faces. In knee-shaped twins there is one plane of symmetry to the combination, and if subsequent twinning bends in the same direction and maintains the plane of symmetry, a cyclic twin develops. This is a very specialized case, but because of its unusual geometry, it has been considered characteristic. A zig-zag type of cyclic twinning which calls for a repetition of a distinctive pattern of alternation is also represented in museum specimens as a characteristic twinning. A more complex type of twinning gives rise to what has been called alphabetical rutile, because the bizarre arrangements often resemble letters of the alphabet. The finest museum specimens of this type represent repeated twinning in which all individuals lie in one plane. The more complex forms of twinned rutile in which twinning occurs randomly in three directions on any of the structurally equivalent (101) faces are usually dismissed as random intergrowths. The exceptional case where three or four successive twinnings have followed the same pattern and the individuals have approximately the same size give rise to distinctive geometric forms which make excellent museum specimens but obscure the structural implications of twinning.

The complexity of twinning in the diamond with its still higher symmetry multiplies so rapidly that a twinned aggregate of more than three individuals nearly defies analysis. Figure 12 is a 14.60 carat gem quality diamond consisting of four octahedrons, three of which are apparent in the photograph. Because the three are not exactly equal in size and because they also partially interpenetrate, it is not at first evident that we are dealing with two octahedrons twinned on a central octahedron. The fourth octahedron, on the underside, is also part of the twinned complex.

Figure 13 illustrated a cyclic twin on (111) of three individuals. *A* and *C* answer all the geometrical conditions of twinning on (114), because it is a common rational plane to which both are symmetrical. If upon continued growth of the aggregate, *A* and *C* became dominant and enclose B, an intergrowth of the type shown in Fig. 14 will develop, which is a form observed among diamond intergrowths.

The possibilities of multiple twinning give rise to such diverse aggregates in the diamond that it is practically impossible to analyze them.

If the individuals of an aggregate are all of the same size, the geometrical symmetry may serve as a basis for analysis. Because the probability of equal size is so remote, the very existence of such highly specialized types leads to the conclusion that irregular twinned aggregates must be common. After examining hundreds of aggregates it does not seem unreasonable to state that most of the intergrowths of diamond commonly classed at random are multiple twins.

ANALYSIS OF TWINNED INTERGROWTHS

When dealing with a twinned intergrowth of three or more individuals, the methods of analytical geometry are much simpler to handle than trigonometric calculations involving interfacial angles. One works with linear equations in three unknowns with coefficients which are small whole numbers, and the algebraic processes are simpler than the complex computations using trigonometric functions which are not rational.

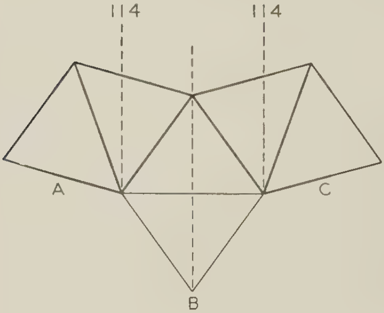


FIG. 13

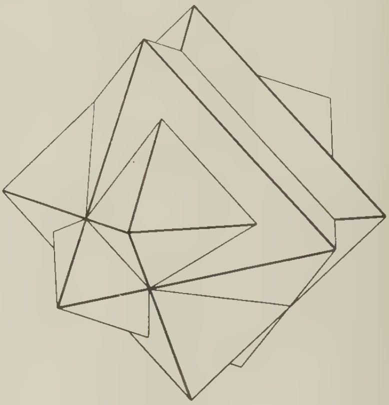


FIG. 14

It is convenient to assume that all planes pass through the origin (Fig. 15) in a manner analogous to the procedures used with the stereographic projection where points are determined by normals to the faces drawn from the center of the reference sphere. If it is desired to find the indices of the new plane (tuv) with respect to the old axes when a given plane (pqr) is reflected through the twinning plane (hkl), the equations of these three planes are written as:

$$px + qy + rz = 0$$
$$hx + ky + lz = 0$$
$$tx + uy + vz = 0$$

By a procedure similar to that by which the reciprocal lattice is derived, the plane (pqr) can be defined by the point p, q, r which will lie on the

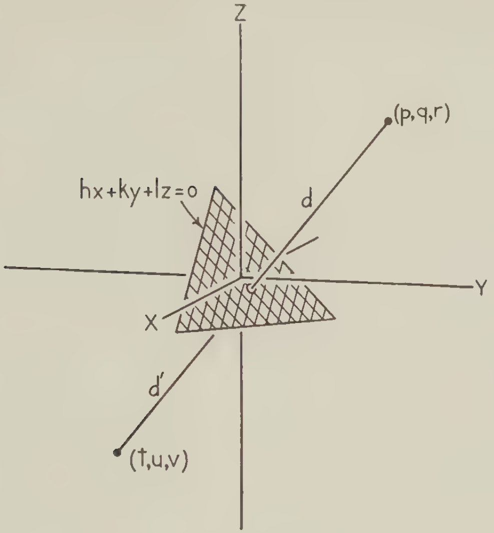


FIG. 15

normal to the plane from the origin. The reflection of that point in (hkl) gives rise to the point t, u, v, which lies on the normal to the plane (tuv) that passes through the origin. The distances of the points p, q, r and t, u, v from the plane $hx+ky+lz=0$ are:

$$d = \frac{hp + kq + lr}{\sqrt{h^2 + k^2 + l^2}} \qquad d' = \frac{ht + ku + lv}{\sqrt{h^2 + k^2 + l^2}}.$$

These distances are equal and opposite in sign, and therefore

$$hp + kq + lr = -ht - ku - lv. \tag{1}$$

If the point t, u, v is the reflection of p, q, r, then t, u, v lies on the line that passes through p, q, r and is normal to $hx+ky+lz=0$. The equations of this line in symmetric form are:

$$\frac{x-p}{h} = \frac{y-q}{k} = \frac{z-r}{l}$$

from which we derive

$$kx - kp = hy - hq \quad \text{and} \quad lx - lp = hz - hr.$$

Because the point t, u, v lies on this line we may substitute t, u, and v for x, y, and z giving:

(2) $kt - kp = hu - hq$ and (3) $lt - lp = hv - hr$.

Rearranging equations (1), (2), and (3) we have

$$\begin{aligned} ht + ku + lv &= -hp - kq - lr \\ kt - hu &= kp - hq \\ lt - hv &= lp - hr. \end{aligned}$$

By substituting the indices of a plane (*pqr*) and the indices of the twinning plane (*hkl*), we have three simultaneous equations which may be solved for *t*, *u*, and *v*. These are actually coordinates of a point, but they also represent the plane (*tuv*). Because (*pqr*) and (*hkl*) are rational (*tuv*) is obviously also rational. In dealing with a twinned crystal, it is therefore possible to use a single set of crystallographic axes and all planes of both individuals will have rational indices on these axes. If a third individual is twinned with the second (Fig. 12), this twinning plane may be referred to the axes of the first and will have rational indices. Then following the procedure above all planes of the third individual can be given rational indices with respect to the axes of the first.

In general: *In any twinning complex of two or more individuals, the crystallographic axes of one individual may be taken as reference axes for all individuals and all planes of the twinned complex will have rational indices with respect to these axes.* In Table I, the forms resulting from twinning of the octahedron, the cube, and the dodecahedron planes on (111), $(\bar{1}11)$ $(\bar{1}\bar{1}1)$, and $(1\bar{1}1)$ are given in the third column. Structurally these represent all the twinning possibilities, because $(\bar{1}\bar{1}\bar{1})$, $(1\bar{1}\bar{1})$, $(11\bar{1})$, and $(11\bar{1})$ are equivalent planes. However, geometrically a completely twinned octahedron could be surrounded by eight octahedrons which structurally represent only four equivalent pairs. In this sense an octahedron twinned on (111) and $(1\bar{1}1)$ is identical structurally with one twinned on (111) and $(\bar{1}11)$, but the physical appearance and geometry of the two groupings are different. In the first instance the two outer octahedrons would be symmetrical to (100) of the central octahedron and in the second symmetrical to (011). This apparent anomaly arises only when one attempts to interpret twinning in terms of the geometry of the aggregates. It again emphasizes the fact that twinning is not a geometric process but a deviation in the process of crystal growth related to the structure of the crystal.

The results of a second twinning of the octahedron only are given in the fifth column of Table II. The rapid increases in the number of possible twinnings demonstrates the degree of complexity which is introduced by multiple twinning. The continued appearance of certain integers in interchanged positions due to the high symmetry is readily apparent.

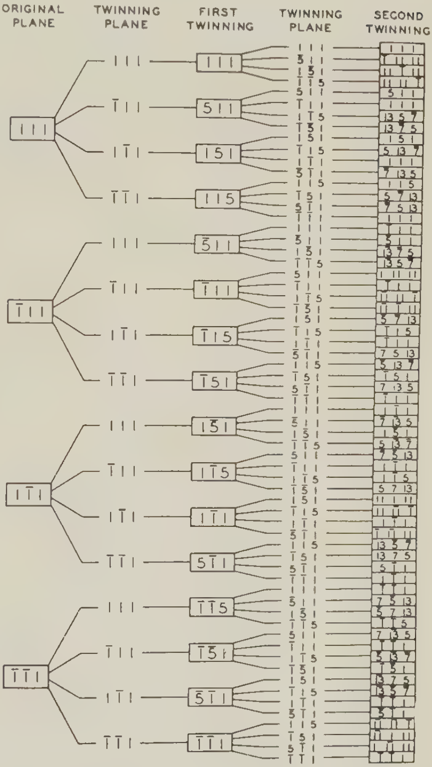
Inclusions and Twinning

If a twinned crystal of diamond is included within another crystal (Figs. 8 and 9), the contacting surfaces will consist of parallel planes (the twinning planes) and vacillating juncture surfaces which, because of their irregular character, may be called suture contacts to differentiate them from twinning contacts. The twinning junctures may be offset as

TABLE I

ORIGINAL PLANE	TWINNING PLANE	FIRST TWINNING
100	111	$1\bar{2}\bar{2}$
	$\bar{1}11$	122
	$1\bar{1}1$	$12\bar{2}$
	$\bar{1}\bar{1}1$	$1\bar{2}2$
010	111	$\bar{2}1\bar{2}$
	$\bar{1}11$	$21\bar{2}$
	$1\bar{1}1$	212
	$\bar{1}\bar{1}1$	$\bar{2}12$
001	111	$\bar{2}\bar{2}1$
	$\bar{1}11$	$2\bar{2}1$
	$1\bar{1}1$	$\bar{2}21$
	$\bar{1}\bar{1}1$	221
110	111	$\bar{1}\bar{1}4$
	$\bar{1}11$	110
	$1\bar{1}1$	110
	$\bar{1}\bar{1}1$	$\bar{1}\bar{1}4$
101	111	$\bar{1}4\bar{1}$
	$\bar{1}11$	101
	$1\bar{1}1$	$\bar{1}4\bar{1}$
	$\bar{1}\bar{1}1$	101
011	111	$\bar{4}\bar{1}\bar{1}$
	$\bar{1}11$	$4\bar{1}\bar{1}$
	$1\bar{1}1$	011
	$\bar{1}\bar{1}1$	011
$1\bar{1}0$	111	$1\bar{1}0$
	$\bar{1}11$	$\bar{1}14$
	$1\bar{1}1$	$\bar{1}1\bar{4}$
	$\bar{1}\bar{1}1$	$1\bar{1}0$
$10\bar{1}$	111	$10\bar{1}$
	$\bar{1}11$	$\bar{1}41$
	$1\bar{1}1$	$10\bar{1}$
	$\bar{1}\bar{1}1$	$\bar{1}\bar{4}1$
$01\bar{1}$	111	$01\bar{1}$
	$\bar{1}11$	$01\bar{1}$
	$1\bar{1}1$	$4\bar{1}1$
	$\bar{1}\bar{1}1$	$\bar{4}\bar{1}1$

TABLE II



in Fig. 8, but they still retain their parallelism, and their traces on a polished surface can be readily distinguished from the suture traces by their regularity. From the foregoing tables it can be seen that the non-twinning boundaries between the two individuals might consist of rational planes with unlike indices in contact with each other, but usually in the diamond the juncture is between step-like faces of either the octahedron or dodecahedron along which each individual is trying to maintain its dominant habit. They resemble closely the type of boundary that exists between two crystals of a random intergrowth. It is commonly the locus of inclusions and open spaces of microscopic size.

A corollary is that the presence of inclusions is indicative of twinning in the adjacent area. Diamond cutters have observed and commented on the fact that inclusions are apt to indicate an area of hardness. It has been postulated that strain in the vicinity of any inclusion has given the diamond a greater hardness at that point, but the logical answer is that the presence of twinning gives an area that presents a harder direction of the diamond to the lap. The high correlation between inclusions and twinning indicates that the diamond cutters term macle for those diamonds which the mineralogist calls twins probably is a corruption of the French word *macle*, meaning spot or blemish.

In many crystals, for example quartz, internal phantom crystals are outlined by inclusions which lie on crystallographic faces representing an earlier growth level of the crystal. It is characteristic of the diamond that when it contains clouds of minute inclusions they do not lie along crystallographic directions but follow the irregular vacillating planes that characterize suture contacts between two twinned individuals.

Conclusions

The examination of large numbers of diamonds indicates that twinning is a dominant characteristic of the diamond. The variation in hardness with direction can be utilized to reveal the twinning which is not observed in polarized light, the common method by which microscopic twinning is observed. The structure of the diamond is such that twinning on (111) should be expected as a normal deviation in the growth process. Because of the high symmetry, twinning usually occurs on more than one of the octahedral faces at the same time. This gives rise to multiple twins of complex nature which have commonly been considered random intergrowths. Inclusions within the diamond are correlated with twinning.

Acknowledgments

This work was carried on with the assistance of grants from the Faculty Research Fund of the University of Michigan and from the Office of Naval Research. Mr. Jack Levy of Detroit did a considerable portion of the experimental cutting in the early phases of the work and has generously contributed with his practical advice toward the solution of technical problems.

THERMAL ANALYSIS STUDIES ON CARBONATES
I. ARAGONITE AND CALCITE[1]

GEORGE T. FAUST, *U. S. Geological Survey, Washington, D. C.*

TABLE OF CONTENTS

Abstract 207
Introduction 207
Description and chemical composition 208
  (a) Aragonite 208
  (b) Calcite 209
  (c) Aragonite-calcite mixture 209
Thermal analysis 209
  (a) Static method 209
  (b) Dynamic method—differential thermal analysis 212
    (1) General statement 212
    (2) Aragonite curves 212
    (3) Calcite curves 215
    (4) Aragonite with calcite curves 215
    (5) Measurements of areas under the thermal curves 219
The aragonite→calcite transformation 221
The formation of unstable polymorphs 222
Acknowledgments 223
References 223

ABSTRACT

Differential thermal analysis studies were made on eight different samples of aragonite, seven of calcite, and one natural mixture of aragonite and calcite. The transformation of aragonite to calcite gave a thermal effect that was easily observed with the continuous-photographic-recording type of differential thermal analysis apparatus. This transformation serves to identify aragonite. The aragonite-calcite transformation and the formation of unstable polymorphs of $CaCO_3$ are discussed.

INTRODUCTION

The writer started his studies on the thermal analysis of carbonate minerals at the U. S. Department of Agriculture in 1942. These data formed the basis of a method for differentiating magnesite and dolomite which was later developed at the U. S. Geological Survey (Faust, 1944). A comprehensive study by thermal analysis of the carbonates and of the hydrous magnesium silicates which was begun in 1944 is now well under way; this paper is the first of a series to be published on studies on carbonate minerals.

A preliminary note regarding the use of the differential thermal analysis method for differentiating between aragonite and calcite was recently published (Faust, 1949).

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DESCRIPTION AND CHEMICAL COMPOSITION OF THE MINERALS

Some of the specimens used in this study were obtained from the U. S. National Museum through the cooperation of Dr. W. F. Foshag and E. P. Henderson, and some were supplied by Drs. C. S. Ross and W. T. Schaller of the U. S. Geological Survey. Some of the specimens were collected by the writer.

The samples used in this study were all carefully selected by hand picking under the binocular microscope. This selected material was then crushed to pass a 58-mesh cloth sieve and re-examined for impurities. The samples were then examined in immersion liquids under the petrographic microscope. Spectrographic analysis of the material and *x*-ray diffraction studies served to verify the purity and the identitity of the minerals.

(*a*) *Aragonite*

ARAGONITE #9. Locality: Chile, South America. (U.S.N.M.—R2554) Labelled cupriferous aragonite. Very pale greenish-blue aggregates. Spectrographic analysis shows 0.01 per cent CuO, 0.12 per cent ZnO, and 0.09 per cent SrO.

ARAGONITE #3 (Zeyringite). Locality: Zeyring, Flatschach, Styria, Austria. (U.S.N.M. #R2548) Green to white masses. This mineral was supposedly nickel-bearing, but spectrographic analysis fails to show nickel as a constituent. The color is probably due to the copper. The chief impurity is strontium, 0.18 per cent SrO corresponding to 0.26 per cent $SrCO_3$. Strunz (1941) states that zeyringite is partly saddle-shaped calcite and partly the variegated variety of aragonite, flos ferri.

ARAGONITE #5. Locality: Enos-Lindsey Cove, Batesville district, Arkansas. White to colorless radiating needles. This is a strontian-barian aragonite containing 0.86 per cent SrO and 0.56 per cent BaO, corresponding to 1.23 per cent $SrCO_3$ and 0.72 per cent $BaCO_3$.

ARAGONITE #6 (Nicholsonite). Locality: Robert Emmett mine, Leadville, Colorado (U.S.N.M. #R2550). White radiating blades arranged in asteriated forms. This variety of aragonite was never fully described. Butler (1913) reports up to 12 per cent zinc. Spectrographic analysis shows the chief impurity of the specimen studied in this investigation to be 0.19 per cent SrO, and no zinc was found.

ARAGONITE #15 (PLUMBOAN). Locality: Mine la Motte, 5–6 miles northwest of Fredericks-town, Madison County, Missouri. Colorless to white radiating needles. Spectrographic analysis shows 1.8 per cent PbO (equivalent to 2.15 per cent $PbCO_3$) and 0.04 per cent SrO.

ARAGONITE #12. Locality: Fifty miles southeast of Roswell and 17 miles east of Lake Arthur New Mexico. Pseudohexagonal cyclic twins. Some of the crystals have a surficial coating of iron oxides. Only the clear cores were used in this study and the success of the separation of the coated grains from the core is attested to by the spectrographic analysis, Table 1, which shows only 0.005 per cent FeO. This aragonite is a strontian aragonite containing 0.73 per cent SrO, equivalent to 1.04 per cent $SrCO_3$.

ARAGONITE #4 (PLUMBOAN) (Tarnowitzite). Locality: Friedrich mine near Tarnowitz, Silesia. (U.S.N.M. #C-2101). Greenish, needlelike crystals of the so-called "grüner stengeliger" variety. Spectrographic analysis shows 3.5 per cent PbO, equivalent to 4.19 per cent $PbCO_3$, and 0.09 per cent SrO. A wide variation in composition for this mineral from the

Friedrich mine is reported in the literature. Doelter (1912) lists an analysis by Traube of the grüner stengeliger variety which shows 4.26 per cent PbO (5.09 per cent $PbCO_3$) and 0.25 per cent SrO.

Aragonite #10 (Mossottite). (U.S.N.M. #R2547). Locality: Lias of Gerfalco in Tuscany, Italy. Pale greenish-blue radiating masses. Spectrographic analysis shows 0.18 per cent SrO, 0.10 per cent PbO, and 0.13 per cent ZnO. Doelter (1912) lists an analysis by Lucca of material from this same locality which contains 6.68 per cent $SrCO_3$, 1.21 per cent $CuCO_3$, 0.82 per cent Fe_2O_3.

(*b*) *Calcite*

Calcite #2. Locality: Ala-Mar deposit, Currant Creek, Nye County, Nevada. White fine-grained masses containing about 10 per cent of admixed dolomite.

Calcite #13. Locality: Joplin district, Missouri. Colorless cleavage fragments. This calcite was analyzed by Dr. R. C. Wells (1915) and found to contain SiO_2, 0.09 per cent; MnO, 0.05 per cent; FeO, 0.19 per cent; CaO, 55.80 per cent; MgO, 0.00; CO_2 (calc'd.), 43.99 per cent; total 100.12 per cent. A comparison with the spectrographic analysis by Murata shows Wells' FeO to be too high and his MgO figure to be in error.

Calcite #F1102. Locality: Franklin Furnace, New Jersey. Orange cleavage masses exhibiting coarse polysynthetic twinning lamellae.

Calcite #11. Locality: Soda Dam Springs, two miles north of Jemez Hot Springs, Sandoval County, New Mexico. White, radiating columns, paramorphous after aragonite.

Calcite #7, (Schaumkalk). Locality: Myokenzan, Harima, Japan. (U.S.N.M. #61, 492). Snow-white scaly masses, paramorphous after aragonite. Spectrographic analysis shows 1.2 per cent MnO.

Calcite #A-3. Locality: Highway cut, one-half mile northeast of Myersville, Maryland. White cleavage masses, showing polysynthetic twinning. See figure I-c in Faust (1944).

Calcite #18. Locality: Champion Mica mine, Amelia County, Virginia. Vug filling.

(c) *Aragonite-calcite mixture*

Aragonite-Calcite #8. Locality: Monte Neva (formerly Melvin) Hot Springs SW$\frac{1}{4}$SW$\frac{1}{4}$ sec. 24, T.21N. R.63E., 18 miles north of McGill, White Pine County, Nevada. A natural mixture. Spectrographic analysis shows MgO to be the chief impurity.

Complete spectrographic analyses for the minor elements present were kindly made by K. J. Murata of the U. S. Geological Survey and are given in Table 1. It is noteworthy that strontium was noted in all the specimens and barium in most of them.

Thermal Analysis

(*a*) *Static method*

The static method of thermal analysis was developed many years ago by mineralogical chemists for the analysis of hydrated minerals. The earliest method of study, and one still used today, consists of heating the sample at a given temperature, cooling in a desiccator, weighing, and then repeating the heating at this temperature until the sample attains constancy in weight. The process is then repeated at other temperatures. The results are commonly plotted as loss in weight of sample

TABLE 1. SPECTROGRAPHIC ANALYSIS OF SOME ARAGONITE AND CALCITE SPECIMENS

K. J. Murata, analyst

Description	Number	MgO	MnO	FeO	SrO	BaO	PbO	ZnO	CuO	NiO	Y_2O_3	V_2O_5	Ag
Aragonite. Chile, South America	9	X	X	0.002	0.09	0.001	0.007	0.12	0.01	X	X	X	X
Aragonite. Styria, Austria	3	0.02	X	0.004	0.18	0.005	X	X	0.04	X	X	X	X
Aragonite. Batesville district, Arkansas	5	0.004	0.002	0.02	0.86	0.56	X	X	X	X	0.006	X	X
Aragonite. Leadville, Colorado	6	0.05	0.001	0.003	0.19	0.02	0.004	X	X	X	X	X	X
Aragonite. Mine La Motte, Missouri	15	0.004	X	0.002	0.04	0.001	1.8	X	X	X	X	X	X
Aragonite. Near Roswell, New Mexico	12	0.01	X	0.005	0.73	X	X	X	X	X	X	X	X
Aragonite. Tarnowitz, Silesia	4	0.009	X	0.003	0.09	0.008	3.5	X	X	X	X	X	X
Aragonite. Tuscany, Italy	10	X	X	X	0.18	0.005	0.10	0.13	0.01	X	X	X	X
Calcite. Currant Creek, Nye County, Nevada	2	2.2	0.04	0.13	0.01	0.003	X	X	X	0.002	X	0.01	X
Calcite. Joplin, Missouri,	13	0.12	0.07	0.10	0.01	X	X	X	X	X	0.005	X	0.0005
Calcite. Jemez Hot Springs, New Mexico	11	0.29	0.15	0.03	0.03	0.004	X	X	X	X	X	X	X
Calcite. Myokenzan, Harima, Japan	7	0.001	1.2	0.08	0.06	X	X	X	X	X	0.008	X	X
Aragonite-calcite mixture. Monte Neva, Hot Springs, Nevada	8	1.5	0.005	0.03	0.02	0.05	X	X	X	X	X	X	X

X, element looked for but not found.

Other elements not found: Sn As Sb Bi Mo W Tl Ge In Co Cd La Cb Ti Zr.

versus temperature. Various improvements in this technique were added from time to time and the apparatus designed and used by Dr. P. G. Nutting (1943), of the U. S. Geological Survey is typical. In this apparatus, the crucible is suspended in the furnace from the balance, which permits the sample to be weighed while in the furnace and eliminates errors introduced by removing the crucible from the furnace.

The static method involves only the determination of the weight of the sample and the measurement of the temperature. It can, therefore, reveal only phenomena associated with a loss or gain in weight. Changes in weight are involved in such reactions as the loss of water or carbon dioxide, and in the oxidation of iron or manganese. The static method cannot detect inversions, for such phenomena are not accompanied by changes in weight.

Dr. Nutting (1948) very kindly examined for the writer by this method two specimens—a calcite from Joplin, Missouri (#13) and aragonite from Enos-Lindsey Cove, Batesville district, Arkansas. The results are given in Fig. 1; they are essentially identical. The temperature of decomposition for both is 615° C.

In the static method of thermal analysis as described here, the process of dissociation is not reversible because carbon dioxide is lost to the atmosphere. The reaction is written:

$$[CaCO_3 + \Delta \rightarrow CaO + CO_2\uparrow]$$

the single horizontal arrow implying thermodynamic irreversibility. The

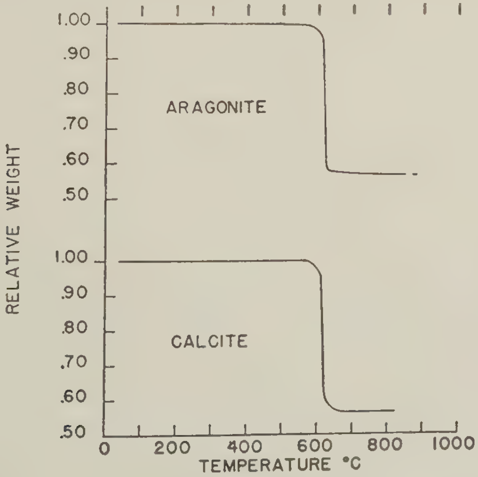

FIG. 1. Thermal analysis curves of aragonite and calcite obtained by the static method. The ordinate is the weight of the sample at each temperature divided by the original weight taken.

temperature of dissociation is, therefore, not an equilibrium value and may vary, depending on such factors as grain size, intergranular texture, presence of impurities, etc.

The data in Fig. 1 are based on relative weight and show a constancy in weight at 0.56. Pure calcite and aragonite contain 56.08 per cent CaO, 43.92 per cent CO_2. The data in Fig. 1 are in accord with this theoretical value and hence the samples were completely dissociated.

The static method is unable to differentiate between calcite and aragonite as it does not reveal the thermal effect that accompanies the transition.

(*b*) *Dynamic method—differential thermal analysis*

(1) General statement

A brief description of the technique employed in this laboratory in making differential thermal analyses and references to the original literature on the apparatus and method are given in a previous paper (Faust, 1948). The curves are produced by the reflection of a light beam from the galvanometer mirrors upon sensitive photographic paper and represent continuous, unhampered recording. The rate of heating for the samples was approximately 12° C. per minute; a complete run required about 80 minutes.

(2) Aragonite curves

The thermal analysis curves for aragonite are given in Fig. 2 and the data are summarized in Table 2. The first four curves C-486, C-484, C-471, C-131 show clear-cut relationships free from complications. In each sample the transition from aragonite to calcite, when it starts, takes place promptly, and the curves show well-defined breaks. In the sample of aragonite from Arkansas (curve C-131) the break is flattened, and this arises from the slightly larger spread in the temperature range over which the transition takes place. The slight hump before the transition break, which appears in this and other records here included, is the result of an accidental instrumental effect and has nothing to do with the aragonite. The straight portions of the curve before and after transition are not noticeably offset, indicating that in this temperature range these two polymorphic forms of $CaCO_3$ have nearly identical specific heats. In the curves showing the inversion of quartz from the low to the high form, the offset in the height of the curves is easily observed (Faust, 1948).

The subsequent dissociation of the calcite paramorphous after aragonite is accompanied by a very large thermal effect. As mentioned previously, the loss of carbon dioxide is approximately 44 per cent of the sample weight. The next six curves, although they show normal relationships

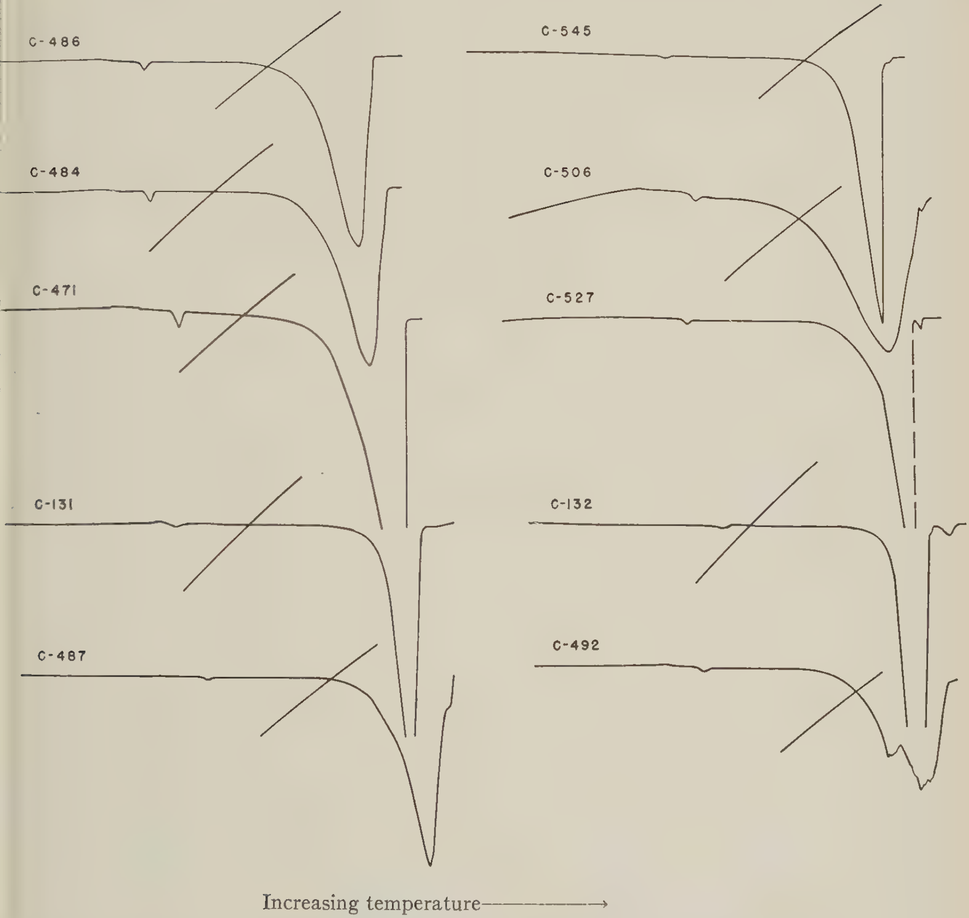

FIG. 2. Differential thermal analysis curves for aragonite. C-486 = aragonite #9, C-484 = aragonite #3, C-471 = aragonite #3, C-131 = aragonite #5, C-487 = aragonite #6, C-545 = aragonite #15, C-506 = aragonite #12, C-527 = aragonite #4, C-132 = aragonite #15, C-492 = aragonite #10. The diagonal line intersecting the curves is a portion of the curve registering the temperature.

TABLE 2. OBSERVATIONS ON THE THERMAL CURVES OF ARAGONITE

Record No.	Sample	Temperature of transition—T_1	Character of break	Temperature of dissociation—T_2	Character of break	Temperature of subsidiary break—T_3	T_2–T_1	T_3–T_2	Locality	Resistance in the galvanometer circuit
C-486	9	447° C.	Sharp	897° C.	Sharp		450° C.		Chile, S.A.	999.9 ohms.
C-484	3	458	Sharp	914	Sharp		456		Flatschach, Styria, Austria,	999.9
C-471	3	462	Sharp	929	Sharp		467		Flatschach, Styria, Austria,	600
C-131	5	400	Rounded	955	Very sharp		555		Batesville district, Arkansas	999.9
C-487	6	461	Rounded	918	Sharp	956 °C.	457	38° C.	Leadville, Colorado	999.9
C-545	15	477	Rounded	943	Very sharp	1156	466	213	Mine La Motte, Missouri	999.9
C-506	12	488	Rounded	890	Rounded	961	402	71	Near Roswell, New Mexico	600
C-527	4	387	Rounded	891	Sharp		504		Tarnowitz, Silesia	600
C-132	15	484	Rounded	959	Very sharp	1019	475	60	Mine La Motte, Missouri	999.9
C-492	10	447	Rounded	870 927 950					Tuscany, Italy	600

at the transition, are complicated at the close of the dissociation of the paramorphic calcite by additional breaks in the curve. These breaks in the curve range from the inflexional change in C-487 and C-545 to the formation of subsequent independent breaks which, for some samples, become more widely separated from the main break, as is shown in records C-506, C-527 and C-132. The sample of aragonite (curve C-492) represents an extreme in that it shows both inflexional alterations of the curve and many subsidiary breaks. The probable origin of these subsidiary breaks will be considered with the discussion of the aragonite-calcite mixtures.

A summary of the observations on these curves is given in Table 2. The temperature of the transition from aragonite to calcite is variable and no relation between this temperature and the content of substituents such as Sr and Pb could be established. The variability of the transition temperature, T_1, and the dissociation temperature, T_2, for duplicate runs is shown clearly in curves C-484 and C-471 obtained with aragonite from Styria, Austria, and C-545 and C-132, aragonite from Mine La Motte, Missouri. These reactions are not reversible and many factors contribute to departures from definite agreement between duplicate runs.

(3) Calcite curves

Thermal analysis curves for calcite are given in Fig. 3, and the data obtained from the curves are summarized in Table 3. All the curves are similar. They all exhibit sharp thermal dissociation breaks. The size of the particles affects the temperature at which dissociation begins, for finer sizes tend to broaden the base of the break.

Theoretically we would expect that the finer particles would cause a spreading of the curve through the onset of dissociation at a lower temperature. Small particles have a much greater surface area than large particles and hence a proportionately greater number of broken bonds. Broken bonds reduce the thermal stability of solids in the boundary layer. As the particles become finer and finer, the importance of the boundary layer increases. Smaller particles also have a greater vapor pressure than larger particles. None of the samples discussed here contained a significant proportion of exceedingly fine particles. It is likely that such fine particles would spread the base of the curve more than was observed.

Twinning of calcite apparently has little if any effect on the character of the thermal analysis curve.

(4) Aragonite with calcite curves

A natural mixture of calcite and aragonite was fortunately available

TABLE 3. OBSERVATIONS ON THE THERMAL CURVES OF CALCITE

Record No.	Sample No.	Temperature of dissociation	Character of the break	Comments	Locality	Resistance in the galvanometer circuit
C-148	2	939° C.	Sharp	Contains some dolomite. (Magnesium is not in the calcite)	Ala-Mar magnesite deposit, Sec. 34 T.12N. R.59E. Nye County Nevada.	600 ohms.
C-89	13	972	Sharp	Analyzed sample of R. C. Wells	Joplin, Missouri	999.9
C-558	F-1102	967	Sharp	Coarsely twinned	Franklin Furnace, New Jersey	999.9
C-552	13	956	Sharp	Analyzed sample of R. C. Wells (−48+65 mesh) = 34% (−200 mesh) = 66%	Joplin, Missouri	600
C-521	13	954	Sharp	Analyzed sample of R. C. Wells (−20+48 mesh) = 24% (−66 mesh) = 76%	Joplin, Missouri	600
C-533	11	937	Sharp	Paramorph after aragonite	Soda Dam Hot Spring deposit, Jemez Springs, New Mexico.	999.9
C-485	7	939	Sharp	Paramorph after aragonite (Scales)	Var. Schaumkalk. Myokenzan Harima, Japan	999.9
F-1*	A-3	952	Sharp	Coarsely twinned.	Highway cut, one-half mile N.E. of Myersville, Maryland.	900
C-307*	18	914	Sharp	Contains pyrite and sericite. Vug filling.	Champion mica mine, Amelia County Virginia. Record by W. Griffitts.	600

* These curves are not given in Fig. 3. The curve for F-1 is given in Faust (1944) on p. 144.

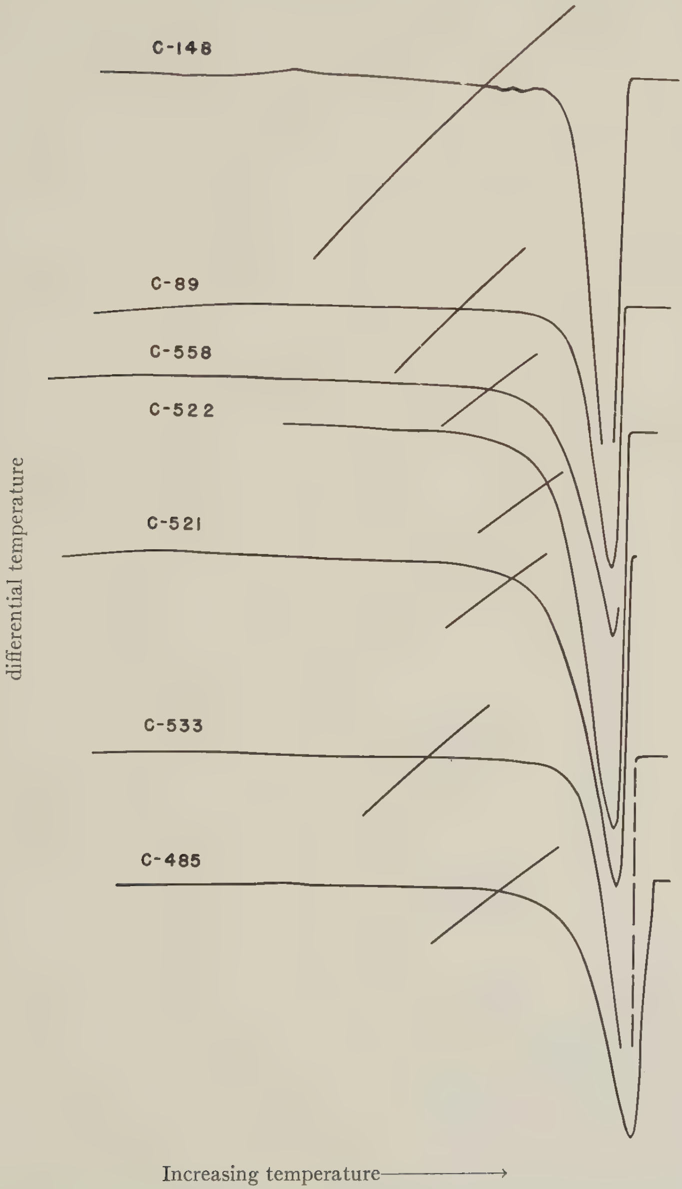

FIG. 3. Thermal analysis curves for calcite. C-148 = calcite #2, C-89 = calcite #13, C-558 = calcite #F-1102, C-522 = calcite #13, C-521 = calcite #13, C-533 = calcite #11, C-485 = calcite #7. The diagonal line intersecting the curves is a portion of the curve registering the temperature.

for study. This material, a spring-water deposit from Monte Neva Hot Springs, Nevada (sample #8), consists of prismatic crystals of aragonite and equant rhombs of calcite. The temperature of the springs that deposited these minerals ranges from 173° F. (78.3° C.) to 193° F. (89.4° C). The thermal curve, C-504, (Fig. 4), shows a small but well-formed peak that corresponds to the transition of aragonite to calcite. The dissociation curve of the calcite is sharp.

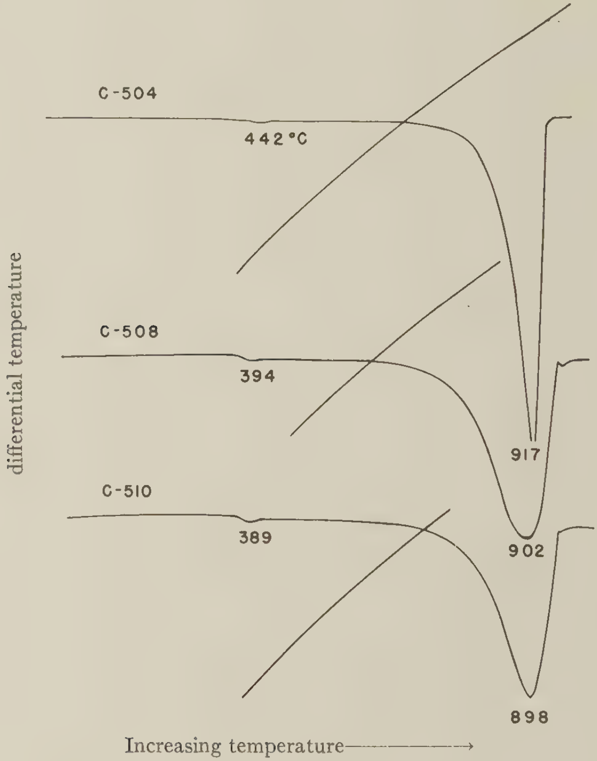

FIG. 4. Differential thermal analysis curves for natural and artificial mixtures of aragonite and calcite. C-504 = natural aragonite-calcite mixture #8, C-508 = mixture of 50% aragonite #5 and 50% calcite #13, C-510 = 90% aragonite #5 and 10% calcite #13. The diagonal line intersecting the curves is a portion of the curve registering the temperature.

In addition to this natural mixture, two artificial mixtures were prepared. The sample used to obtain curve C-508 was made up of 50 per cent of aragonite #5 and 50 per cent of calcite #13, whereas that for curve C-510 was made up of the same minerals in the proportions of 90 per cent aragonite to 10 per cent calcite. Both of the minerals used in the mixtures give normal uncomplicated patterns. The curves, C-508 and C-510, both show the transition of aragonite to calcite as was anticipated. The break showing the dissociation of calcite is markedly different from that of either of the pure minerals used. This break in the curve is a compound break and arises from the presence of two types of calcite, that which results from the transition of aragonite to calcite and that used in the mixture. Each of these types of calcite dissociates at slightly different temperatures, hence the reactions overlap. The break in curve C-508 due to the dissociation of calcite has a very wide base and a rounded top. The temperature of this break is 902° C., much lower than the mean values of the pure minerals used, namely 963° C. The shifting of the temperature of the break in thermal analysis studies on mixtures has been pointed out by many workers. The temperature of this break for the mixture of 90 per cent aragonite and 10 per cent calcite is even lower than that mentioned previously (898° C.) for curve C-510. A curious subsidiary break, of very small size, occurs in curves for mixtures after the curve for the principal dissociation of calcite. This break does not appear in either of the pure mineral curves and is believed to arise through the delayed dissociation of some of the larger grains of calcite. Measurements and observations on these curves are given in Table 4.

The appearance of the subsidiary breaks on the aragonite curves, mentioned earlier, may arise in part as a consequence of difference in size of the particles. It may also be due to intra-particle texture or to the presence of substances in solid solution not uniformly distributed through the fragments, as in zoned crystals. The presence of small amounts of calcite, not produced by the present rapid inversion of aragonite to calcite in the sample, and hence lacking in the strain induced by paramorphism, could also give rise to subsidiary breaks. It should be noted (Fig. 4) that the curve for 50 per cent aragonite and 50 per cent calcite almost was resolved into two breaks (curve C-508).

(5) Measurements of areas under the thermal curves

Areal measurements were made, using a polar planimeter and checking with a simple grid, on five records, all with 999.9 ohms resistance in the galvanometer circuit. These measurements were repeated at least five times for each curve; the agreement between successive measurements on the same curve was satisfactory. From the data for each curve, the area corresponding to the transformation of 1 gram of aragonite to calcite was calculated. The complete lack of agreement among the calculated results was disappointing. The average area arising through the transformation of 1 gram of aragonite to calcite was found to be 30 square millimeters, the maximum being 43.8 square millimeters and the minimum 17.5 square millimeters.

TABLE 4. OBSERVATIONS ON THE THERMAL CURVES OF MIXTURES OF ARAGONITE AND CALCITE

Record number and description	Sample No.	Temperature of transition–T_1	Character of break	Temperature of dissociation–T_2	Character of break	Temperature of subsidiary break–T_3	T_2–T_1	T_3–T_2	Resistance in the galvanometer circuit
C-504. Aragonite and calcite Monte Neva (formerly Melvin) Hot Springs. SW $\frac{1}{4}$ Sec. 24, T.21.N. R.63E, White Pine County, Nevada.	8	442° C.	Rounded	917° C.	Sharp		475° C.		999.9 ohms
C-508. Aragonite, Batesville District, Arkansas, 50%; Calcite, Joplin District, Missouri, 50%.	5 and 13	394	Rounded	902	Rounded	964° C.	508	62° C.	999.9
C-510. Aragonite, Batesville District Arkansas 90%; Calcite, Joplin District, Missouri 10%.	5 and 13	389	Rounded	898	Sharp	948	509	50	999.9

The heat of transformation of aragonite to calcite is given by Goranson (1942) as $\Delta H = +21$ joules per gram. The positive sign indicates that the reaction is endothermic. The area under the curve, given above, is produced by heat effects of this order of magnitude.

The heat of transformation is given off in the rearrangement of the atoms and groups of atoms in the crystal structure of aragonite as they shift to their new positions in the calcite structure. This rearrangement has been studied by Kôzu and Kani (1934) and the reader is referred to their paper for the details.

The area under the curve for the dissociation of calcite paramorphous after aragonite was also measured for the same series of curves mentioned above. It was found that the dissociation of 1 gram of calcite produces an average area of 5,646 square millimeters, with a maximum of 6,836 square millimeters and a minimum of 4,743 square millimeters. These results apply to curves obtained with 999.9 ohms in the galvanometer circuit. The value of the heat of the reaction, ΔH, for the dissociation of calcite is given by Smyth and Adams (1923) as follows: 9,680 joules at 800° C.; 8,132 joules at 1,000° C.; and 8,654 joules at 1,200° C. The value of ΔH at 898.6° C., the temperature at which $p = 760$ mm. is 9,426 joules. These values of ΔH cover the range of temperatures of 800°–1,200° C. and are computed from data for the system at equilibrium. They indicate the order of magnitude of the heat effect which produces the curves.

The chief difficulty in accurate areal measurement is connected with construction of the straight line used to close the base of the curve. This is further complicated by the steepness of the curve at the onset of transformation. The observations on the measurements of the area under the curve suggest that the use of a simple grid for measurement of the area is entirely adequate.

The variation in the area associated with the transformation of aragonite to calcite imposes a limitation upon quantitative estimates of mixtures of aragonite and calcite. If such estimates are made, it will be necessary to first determine the area under the curve associated with the transformation for the particular aragonite being studied.

THE ARAGONITE→CALCITE TRANSFORMATION

The aragonite→calcite transformation is monotropic in character. It is therefore irreversible and does not take place at a constant temperature.

It has long been well established that merely heating aragonite in air to about 450° C. is sufficient to convert it to calcite. Johnston, Merwin, and Williamson (1916) found that this transformation could be brought about in a few days at temperatures as low as 100° C. by heating in the presence of water. They found that pure dry aragonite was changed to calcite in a few minutes when heated at 470° C., in 90 minutes at 425° C., and in 3 hours at 400° C. Kôzu and Kani (1934) in their studies on the thermal expansion of aragonite crystals observed that the transformation took place, in air, in the range of 450–490° C. More recently, Zimens (1937) using the Hahn radioactive-emanation technique, showed that the transformation of aragonite to calcite begins at about 450° C. These observers all agree on the irreversibility of the aragonite-calcite transformation. The present study shows that at a heating rate of 12° C. per minute, the transformation takes place between 387° and 488° C.

THE FORMATION OF UNSTABLE POLYMORPHS

Credner (1870) made a very thorough study of the synthesis of calcite and aragonite. He found that calcite is precipitated from pure solutions of "doppelt kohlensaurem Kalkes," whereas both calcite and aragonite are precipitated from solutions of calcium bicarbonate that contain limited amounts of lead carbonate, calcium sulfate, or strontium carbonate. He also observed that temperature of precipitation and strength of the solution were not "unique" factors in the cause of the dimorphism of calcium carbonate. This study of Credner is noteworthy, considering the early date of the research. He supported his chemical studies by microscopic examination of the phases and reproduced drawings of the crystals. in his paper. He further tested his conclusion that strontium entered into the aragonite crystals by using a spectroscope to determine its presence. The standard French reference books on mineral synthesis paid no attention to his work and it was soon forgotten. Johnston, Merwin, and Williamson (1916) also observed the formation of aragonite from solutions to which Pb, or sulfate was added.

Bloom and Buerger (1937) have discussed the genesis of polymorphous forms, including the aragonite-calcite relations, and conclude:

> Certain types of impurities present in solution during precipitation of a solid phase often effect the generation of an unstable polymorphous modification. The modification thus generated often contains this impurity, sometimes held so tenaciously as to defy usual chemical treatment for its removal. Under these conditions, the structure containing this impurity shows a remarkable resistance to transformation to the stable structure of the pure substance. The complete removal of the impurity is apparently necessary for the realization of the transformation.

Spectrographic examination showed the presence of barium, strontium, or lead in all of the aragonites used in this study. The total amounts of the oxides of these metals are 0.1 per cent or more. The two specimens of calcite paramorphous after aragonite contain a total of 0.03–0.06 per cent of these oxides. Calcite from Joplin, Missouri, has only 0.01 per cent of these oxides. These results suggest the desirability of further spectrographic studies on these minerals.

TABLE 5. IONIC RADII (IN ANGSTROM UNITS) OF THE CATIONS OF THE CALCITE AND ARAGONITE GROUPS (AFTER BRAGG, 1937)

	Mg	Fe	Zn	Mn	Cd	Ca	Sr	Pb	Ba
Calcite type	0.78	0.83	0.83	0.91	1.03	1.06	—	—	—
Aragonite type	—	—	—	—	—	1.06	1.27	1.32	1.43

The presence of barium, strontium, and lead in solid solution in aragonite is well known. However, zinc does not form an orthorhombic carbonate, and one would anticipate, from consideration of the atomic radii listed in Table 5, that substitution of Ca by Zn would tend to favor the formation of a rhombohedral carbonate. It was of interest, therefore, to re-examine nicholsonite, the reputed zincian aragonite, which Butler claimed (1913), contains as much as 12 per cent zinc. The specimen used in this study was from the Canfield collection of the U. S. National Museum and consists of white radiating blades arranged in asteriated form. Spectrographic analysis failed to reveal determinable quantities of zinc in this sample. The *d* spacings of the powder photograph agree with those of aragonite. Zinc was, however, found in two of the aragonite specimens, which contained 0.12 and 0.13 per cent ZnO. Both of these samples are fine-grained, and neither *x*-ray study nor microscopical observations can unequivocally rule out the possibility of the presence of some minor amount of a zinc mineral. The presence of zinc in solid solution in aragonite thus needs further study.

ACKNOWLEDGMENTS

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THE EFFECT OF THE INTERLAYER CATIONS ON THE EXPANSION OF THE MICA TYPE OF CRYSTAL LATTICE

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Abstract

The ionic radii, the valency, and the total charge of the interlayered cations, as well as the nature of the interlayered substance seem to determine the extent of the interlayer expansion of the mica type of crystal lattice.

Recent investigation of the base exchange properties of vermiculite (2) together with results reported herein, shed considerable light on the factors other than degree of hydration which affect the expansion along the *c* axis of the mica type of a crystal lattice. One of these of special importance is the interlayer cations. The mica type minerals with which this effect has been noted by others or studied experimentally include vermiculite, biotite, montmorillonite, paragonite, muscovite, glauconite, celadonite, hydrous mica clay minerals, and margarite.

Studies with Vermiculite

The effect of the exchangeable ion on the expansion of air dry vermiculite is shown in Table 1. It is seen that the expansion of the lattice saturated with Ba, Li, or Na ions is equivalent to the thickness of a monomolecular layer of water; whereas when saturated with Mg, Ca, or H ions the expansion is equivalent to the thickness of two layers of water molecules. In contrast is the lattice saturated with K, NH_4, Rb, or Cs which is only slightly expanded, the amount being caused apparently by the size of the exchangeable ion (2).

Table 1. Basal Reflections* $d(002)$ for Vermiculite Saturated with Various Cations as Related to Ionic Radius and Charge of Ion

Saturating cation	Mg	Ca	Ba	H	Li	Na	K	NH_4	Rb	Cs
Ionic Radius (13) Å	0.65	0.99	1.35	0.30†	0.60	0.95	1.33	1.48	1.48	1.69
Charge of Ion	2	2	2	1	1	1	1	1	1	1
Basal Reflection $d(002)$ Å	14.33	15.07	12.56	14.33	12.56	12.56	10.42	11.24	11.24	11.97

* This interlayer spacing is usually designated as the $d(002)$ spacing for vermiculite and the micas and as the $d(001)$ spacing for montmorillonite.

† Covalent radius.

It is important to note that although the crystal lattice of Li, Na, or Ba vermiculite expands the equivalent of a monomolecular layer of water when immersed in water (Table 2), the total interlayer expansion of these forms and also of the Mg or Ca forms does not exceed two monomolecular layers of water. The contracted nature of the crystal lattice of K, NH_4, Rb, or Cs vermiculite is further indicated by the fact that no change in the interlayer spacing takes place when immersed in water.

TABLE 2. BASAL REFLECTION $d(002)$ FOR VERMICULITE* SATURATED WITH VARIOUS CATIONS WHEN IMMERSED IN WATER†

Saturating cation	Mg	Ca	Ba	Li	Na	K	NH_4	Rb	Cs
Basal Reflection $d(002)$ Å	14.47	15.41	15.41	15.07	14.76	10.57	11.22	11.22	11.97

* Vermiculite no. 2 (2).

† To obtain x-ray diagrams of material immersed in water, the sealed ends of capillary tubes packed with air dry material were broken off and immersed in water. After the samples became saturated with water by capillarity the two ends of the tubes were sealed with De Khotinsky cement to prevent water loss during exposure to x-rays.

STUDIES WITH MONTMORILLONITE

The effect of the exchangeable ion on expansion of montmorillonite in water has been investigated by Hendricks *et al.* (8) and by Hofmann and Bilke (10). The effect appears in the following manner:

(1) At relatively low states of hydration Mg, Ca, Sr, or H saturated montmorillonite expands to a thickness equivalent to two monomolecular layers of water, whereas Ba, Li, Na, or K saturated forms expand to a thickness equivalent to only one monomolecular layer of water.

(2) Mg, Ca, Sr, or H montmorillonite expands from a thickness of one molecular layer of water to a thickness of two molecular layers of water, at very low states of hydration (3), but Ba, Li, or Na montmorillonite does not expand similarly until the water content is relatively high and sufficient to form a continuous monomolecular layer of water consisting of hexagonal rings of at least two water molecules per half unit cell (3).

(3) The lattice of K or Cs montmorillonite apparently does not expand beyond the equivalent of a monomolecular layer of water even at very high relative humidities.

The effect of the exchangeable ion on expansion of montmorillonite determined in the present investigation is shown in Table 3. It is interesting to note that the interlayer spacing of the air dry samples of Mg, Ca, Ba, H, Li, or Na montmorillonite is approximately the same as with air dry vermiculite saturated with the same ion. However, montmorillonite saturated with K, NH_4, Rb, or Cs ions differs from vermiculite saturated with these ions in that the former have an expanded lattice the equivalent of a monomolecular layer of water, whereas the latter have a non-expanded lattice.

Upon immersion in water, under the conditions specified, montmorillonite saturated with Mg, Ca, Ba, H, Li, or Na ions assumes interlayer

TABLE 3. BASAL REFLECTIONS $d(001)$ FOR AIR DRIED MONTMORILLONITE SATURATED WITH VARIOUS CATIONS AND WHEN IMMERSED* IN WATER

Saturating cation		Mg	Ca	Ba	H	Li	Na	K	NH_4	Rb	Cs
Bentonite‡ No. 2	Air Dry§	14.8	15.1	12.9	14.5	13.4	11.9	12.0	12.1	12.3	12.9
	Immersed in Water‖	20.7	17.9	18.5	19.3	23–18†	24–18†	13.2	13.2	12.3	
Bentonite‡ No. 5	Air Dry		15.2				12.1	11.9			
	Immersed in Water		19.6				24–18	13.2			
Bentonite‡ No. 7	Air Dry	15.1	15.1	12.6			12.6	12.3	12.4		
	Immersed in Water	19.4	19.6	19.6			24–18	13.2	13.7		

* To obtain x-ray diagrams of materials immersed in water, thick pastes were prepared and then packed in capillary tubes. The tubes were sealed with De Khotinsky cement to prevent water loss during exposure to x-rays.

† Li and Na montmorillonites form gels upon wetting; it was found that the expansion varied with the concentration of the gel. The reflection lines were broad, diffused, and very weak in dilute gels; in more concentrated gels the lines were somewhat narrower and with somewhat stronger intensities but still diffused.

‡ See note † Table 4.

§ The relative intensity of the reflection lines according to cation is as follows: $Ca = Mg > Na > Ba = Li = NH_4 > H = K > Rb > Cs$. The intensity of the lines of the last two cations are very weak and diffused.

‖ The relative intensity of the reflection lines according to cation is as follows: $Mg = Ca = Ba > H = NH_4 > K > Na = Li > Rb > Cs$. The lines of Mg, Ca, Ba, and H ions are narrow and with sharp edges, those of NH_4 and K ions are a little broader and with edges which are not as sharp, but the lines of Li, Na, Rb, and Cs ions are very diffused.

spacings equivalent to three, four, or more (possibly in suspensions) monomolecular layers of water (3) (Table 3), whereas the similarly saturated vermiculite samples, do not expand beyond the equivalent of two monomolecular layers of water. K, NH_4, Rb, or Cs montmorillonite, upon immersion in water shows only a relatively small change in the interlayer spacing amounting possibly to an expansion equivalent to one layer of water molecules.

The similarity as well as the difference between montmorillonite and vermiculite saturated with like ions is also clearly demonstrated by the differential thermal curves of the air dried samples (see Figs. 1, 2, and 3, and also the differential thermal curves of the vermiculites previously published (2)). The significant portion of the curves is represented by the first endothermic trough at low temperatures. It is seen that two types of troughs are present, namely, that of the Mg, Ca, Ba, H, or Li montmorillonite and that of Na, K, NH_4, Rb, or Cs montmorillonite. Whereas the bottom of the trough of Na, K, NH_4, Rb, or Cs montmorillonite has one sharp point, the bottom of the trough of Mg, Ca, Ba, H, or Li montmorillonite has two sharp points. The two sharp points suggest, possibly, that these are points of two endothermic troughs, as in the differential thermal curves of the Ca or Mg vermiculite, but which overlapped on account of their close proximity. This is apparently also true of H vermiculites (Fig. 3). The most striking dissimilarity between mont-

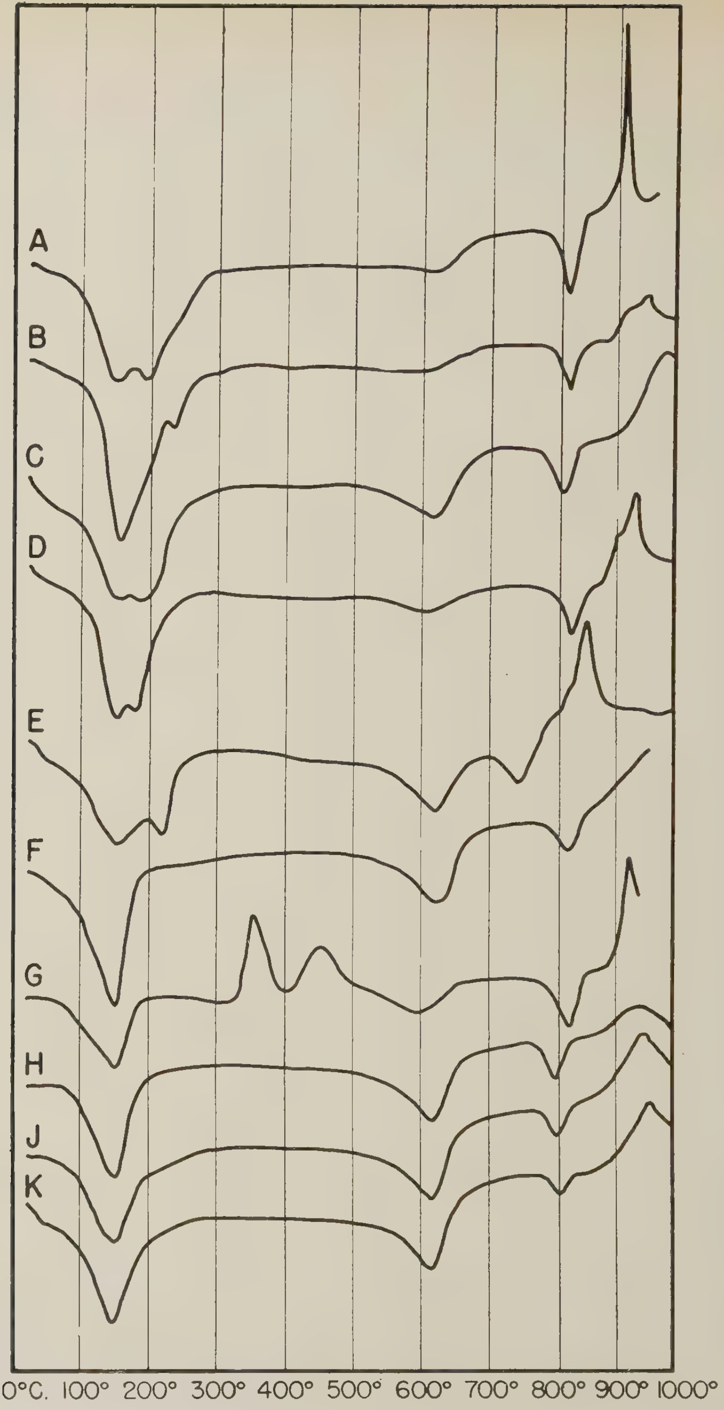

FIG. 1. Differential thermal curves of montmorillonite from bentonite No. 2.

A. Mg saturated
B. Ca saturated
C. Ba saturated
D. H saturated
E. Li saturated
F. Na saturated
G. NH_4 saturated
H. K saturated
J. Rb saturated
K. Cs saturated

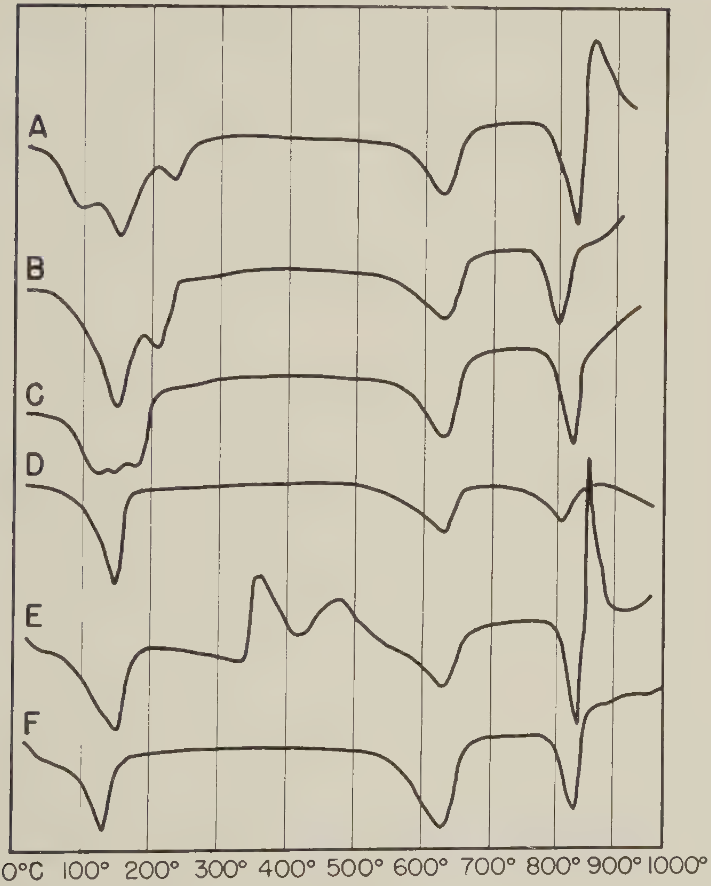

FIG. 2. Differential thermal curves of montmorillonite from bentonite No. 7.

A. Mg saturated	D. Na saturated
B. Ca saturated	E. NH_4 saturated
C. Ba saturated	F. K saturated

morillonite and vermiculite appears in the NH_4, K, Rb, and Cs saturated forms. The former have an endothermic trough at the low temperatures whereas the latter do not. As with vermiculites, the exchangeable (adsorbed) ion influences somewhat the exothermic reaction at ignition temperatures (Figs. 1 and 2).

The differential thermal curve of NH_4 montmorillonite is of special interest, for it has two exothermic breaks, the peaks of which are at 360° C. and 470° C., and these are absent in the curves of the other montmorillonites. These two breaks are apparently associated with the loss of NH_3, for they correspond to the temperatures at which NH_3 is lost as reported by Cornet (6).

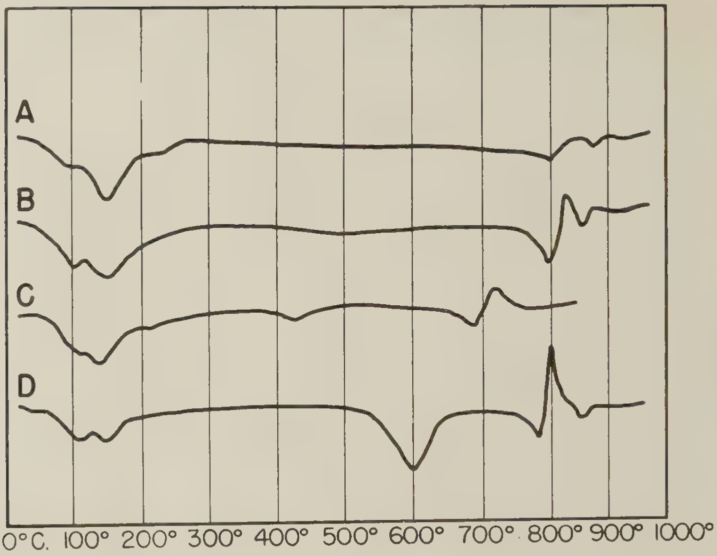

Fig. 3. Differential thermal curves of hydrogen saturated vermiculites (2).

A. Vermiculite No. 1.
B. Vermiculite No. 2.
C. Vermiculite No. 3.
D. Vermiculite No. 5.

Expansion in Glycerol

To clarify further the effect of the interlayer ions on lattice expansion, the interlayer spacing of vermiculites and montmorillonites were determined when immersed in glycerol. MacEwan (11, 12) and Bradley (4) have shown that montmorillonite saturated with either H, Na, NH_4, Ca, or Pb^{++} ions forms a complex with glycerol whereby two monomolecular layers of glycerol occupy the interlayer space. This results in an interlayer spacing of 17.7 Å. However, MacEwan reported that vermiculite does not form a glycerol complex.

In the present study the method employed to saturate vermiculite and montmorillonite with glycerol was as follows: Air dry powdered samples were packed in glass capillary tubes. The sealed end of the tubes were broken off and held in hot (110° C.) water-free glycerol until glycerol rose by capillarity to the top of the sample. Samples of vermiculite #2 (2) and several montmorillonite samples saturated with various ions were thus treated. The results are reported in Table 4.

It is seen that the glycerol treated vermiculite saturated with Mg, Ca, Ba, H, Li, or Na have an identical interlayer spacing of about 14.33 Å. Apparently wetting the Mg saturated form (the natural vermiculite) with glycerol caused no visible change in the interlayer spacing, but the

TABLE 4. BASAL REFLECTIONS FOR VERMICULITE* AND MONTMORILLONITE† SATURATED WITH VARIOUS CATIONS WHEN IMMERSED IN GLYCEROL

Saturation cation	Mg	Ca	Ba	H	Li	Na	K	NH$_4$	Rb	Cs
Sample										
Vermiculite	14.33	14.18	14.33	14.33	14.33	14.33	10.64	11.22	11.22	11.97
Bentonite no. 2‡	17.7	17.7	17.7	17.7	17.7	17.7	14.2	14.5	14.4	15.4
Bentonite no. 1		17.8					14.2	14.5		
Bentonite no. 4		17.8					14.2	17.8		
Bentonite no. 5		18.0					14.4	17.7		
Bentonite no. 7	17.7	17.7	17.7			17.7	14.1	17.7		
Bentonite no. 14		17.7					14.1	17.2		
Bentonite no. 15		17.8					14.1	17.7		
Bentonite no. 16A		17.7					14.1	17.7		
Bentonite no. 16B		17.7					14.1	17.7		
Bentonite no. 17		18.0					14.1	17.7		

* Vermiculite described as no. 2 (2).

† Montmorillonite samples consisting of fractions of $<1\mu$ particles were obtained by sedimentation from the following bentonites:

Bentonite no. 1 from Death Valley, California.
Bentonite no. 2 also called Otaylite from Otay (near San Diego), California.
Bentonite no. 4 from Merrit, British Columbia, Canada.
Bentonite no. 5 from Rosedale, Alberta, Canada.
Bentonite no. 7 from Goldfield, Nevada.
Bentonite no. 14 from Newell, South Dakota.
Bentonite no. 15 from Death Valley, Junction, California.
Bentonite no. 16A from Newberry, California (white colored).
Bentonite no. 16B from Newberry, California (red colored).
Bentonite no. 17 also called Saponite and Hectorite from Hector, California.

‡ The intensity of the reflection is affected by the cation; the relative intensity of the lines according to cations is as follows: $Ca=Mg=Na=H>Ba=NH_4>K>Li>Rb>Cs$. The intensity of the Rb line is very weak and that of Cs is very very weak; the lines are quite diffused. The basal reflection lines of glycerol wet material are narrower and with sharper edges than those of air dry or water saturated material.

change in the other forms was very marked (compare Tables 1 and 4). Thus the Ca form showed a small contraction of the lattice as compared to that of the water treated material while the Ba, Li, or Na forms showed an expansion. These changes indicate that a complex was formed between the vermiculite and glycerol similar to the montmorillonite-glycerol complex but with one monomolecular layer of glycerol instead of two.* It should be noted that a monomolecular layer of glycerol has approximately the same thickness as two layers of water molecules organized in the manner indicated (3). This, perhaps, is the reason for MacEwan's failure to observe the formation of a glycerol complex in the case of vermiculite. K, NH$_4$, Rb, or Cs vermiculite showed no change in the interlayer spacing when wetted with glycerol. This again confirms the results previously observed that the lattice of these minerals is contracted and consequently glycerol is unable to enter the interlayer space of the lattice to bring about expansion.

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* This was verified by a study in which vermiculite samples were dehydrated at 250° C. before treating with hot anhydrous glycerol. The vermiculite glycerol complex formed had nearly the same interlayer spacing as reported in Table 3. Details of this study will be reported at a later date.

The results with montmorillonite confirm those reported by MacEwan. An interlayer spacing of 17.7 Å was obtained not only for H, Na, or Ca saturated montmorillonite but also with Mg, Ba, and Li forms. The interlayer spacing of NH_4 saturated montmorillonites was not consistent; out of ten samples from different sources eight gave the 17.7 Å while two gave a 14.5 Å spacing. These two expanded the equivalent of one layer of glycerol molecules instead of two as in the case of the other forms. The interlayer spacing of all ten of the K saturated montmorillonites was 14.1–14.2 Å. This again is the equivalent of only one layer of glycerol molecules.Although Rb and Cs saturated montmorillonite from Otay, California, gave very weak and diffused lines, the position of the 001 line was approximately that of the K montmorillonite.

The results obtained with glycerol treated NH_4 montmorillonite seem somewhat anomalous. From the similarity of NH_4 and K montmorillonite in their air dry and wet states it would be expected that this similarity might hold true in glycerol. Instead only two out of ten NH_4 montmorillonite samples studied gave spacings similar to that of K montmorillonite. The reason for this is not known.

From the foregoing it may be concluded that when montmorillonite and vermiculite are saturated with like ions, the former expands to a greater extent than the latter in glycerol or water.†

The difference in expansion between vermiculite and montmorillonite is of particular significance when K, NH_4, Rb, or Cs are the adsorbed ions. Vermiculites with these adsorbed cations have contracted lattices whereas montmorillonites have expanded lattices. This difference possibly may explain the near absence of K fixation in montmorillonite and the large fixation exhibited by vermiculite.

As previously indicated (2) fixation or irreversibility of the base exchange reaction in vermiculite occurs between ions which produce a contracted lattice and since no such phenomenon takes place in montmorillonite with the ions studied, irreversibility is also absent.

The Effect of Particle Size on Expansion in Vermiculite

To eliminate the possibility that the observed smaller interlayer expansion of vermiculite as compared with that of montmorillonite was due to the larger dimensions of the vermiculite particles, the expansion of small particles ($<0.25\mu$) was studied. The small particles were obtained by grinding and sedimentation. The results showed that the expansion of these particles whether air dry, immersed in water, or in glycerol was the same as that of larger particles.

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† A similar study with many other organic substances led essentially to the same conclusion. This study will be reported separately.

Studies with Paragonite and Muscovite

Additional evidence for the effect of the interlayer cation on lattice expansion was obtained with paragonite—a mineral similar to muscovite in its composition but with Na as the dominating interlayer ion.

Paragonite samples #1 and #2 studied by Schaller and Stevens (15) were kindly furnished by R. E. Stevens for the present investigation.

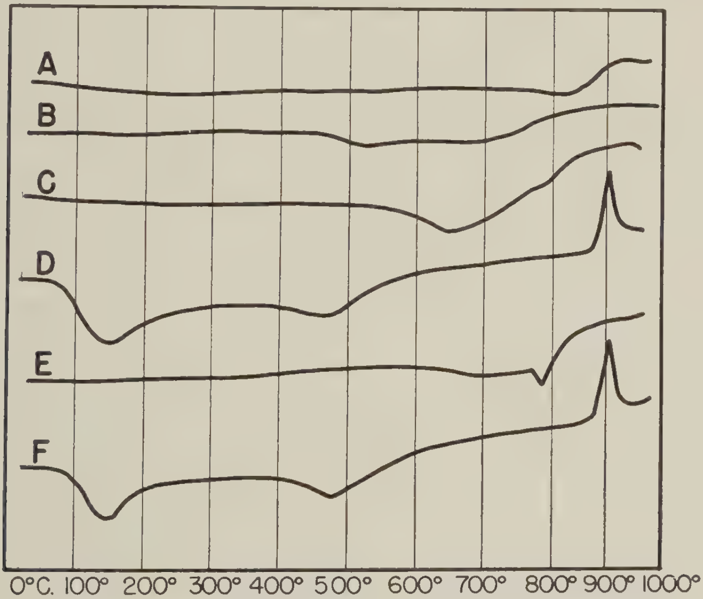

Fig. 4. Differential thermal curves of muscovite and paragonite.

A. Muscovite 5–50μ particles.
B. Muscovite <0.5μ particles
C. Paragonite #1 5–50μ particles.
D. Paragonite #1 <0 5μ particles.
E. Paragonite #2 5–50μ particles.
F. Paragonite #2 <0.5μ particles.

Since paragonite is a true mica formed under high temperatures and pressures, it would be expected to be dehydrated and contracted in its natural form. This was verified by the differential thermal curve (Fig. 4) and the *x*-ray diagrams of these samples (Table 5). The *x*-ray diagrams were identical to those of muscovite with the exception that the basal spacing $d(002)$ is at 9.7 Å instead of 10.0 as for muscovite.

From the results reported herein it was anticipated that if the parag-

TABLE 5. BASAL REFLECTIONS $d(002)$ FOR PARAGONITE* AND MUSCOVITE† FOR COARSE (5–50μ) AND FINE (<0.5μ) PARTICLES‡ WHEN AIR DRY, IMMERSED IN WATER, OR IMMERSED IN GLYCEROL

		Air dry	Immersed in water	Immersed in glycerol
Muscovite	5–50μ	10.06	10.06	10.06
	<0.5μ	10.06	10.06	10.06
Paragonite No. 1	5–50μ	9.7	9.7	9.7
	<0.5μ	12.9 9.7	14.3 9.7	14.3 9.7
Paragonite No. 2	5–50μ	9.7	9.7	9.7
	<0.5μ	12.9 9.7	14.3 9.7	14.3 9.7

* Paragonite No. 1 from Fenestrella, Italy. Paragonite No. 2 from Monte Campiani· Switzerland. These samples were kindly furnished by R. E. Stevens (15).

† Muscovite from Pennington County, South Dakota.

‡ The samples were ground in an agate mortar and fractionated by sedimentation.

onite samples should be ground to extreme fineness, i.e., to less than 0.5 μ, they would hydrate or form the glycerol complex and thereby expand. Muscovite, however, receiving the same treatment, i.e., grinding, should show no such change.

The results of this treatment are shown in Table 5 and Fig. 4. It is seen that the grinding brought about expansion and hydration in the case of paragonite but not with muscovite. These results leave no doubt that the Na ion imparts to the paragonite, when in extreme fineness, the ability to hydrate and expand. It is to be expected that even coarser fractions would hydrate if given enough time.

GLAUCONITE AND CELADONITE

The relation between H_2O content and K_2O, Na_2O and CaO of glauconite and celadonite, reported by Hendricks and Ross (9), indicates that the hydration of these micaceous minerals is also affected by the nature of the interlayer cation. In nearly all the samples the water content found was greater than the theoretical OH^- ions of the framework, namely, about 5 per cent. Nearly all the samples contain some Na_2O and CaO but those samples which contain the largest amounts of Na_2O and CaO and the least amounts of K_2O contain also the largest amounts of water.

STUDIES WITH HYDROUS MICA

The clay minerals resulting from the weathering of micas are known by a variety of names such as "illite," "hydrous mica," etc. (14). All contain water of hydration. It was not realized until the present study that the hydration of the micas upon weathering was mainly the consequence of the replacement of the K by Ca, Mg, and H ions. Hendricks and Alexander (7) state, "In particular, montmorillonite and hydrous micas probably occur together." Similarly Aldrich *et al.* (1) state, "Some soil clays which earlier had been characterized as being principally hydrous mica were shown under these conditions of preparation to consist largely of montmorillonite." The criteria for montmorillonite used by these investigators was based solely on the expansion of the crystal lattice.

The results, however, of the present investigation have shown that interlayer lattice expansion is not confined to montmorillonite but it is a property of the mica type of a crystal lattice and is dependent upon the interlayer cation. For this reason the clay mineral with the expanded lattice reported to be associated with the "hydrous mica" is not necessarily montmorillonite as such, but is probably the result of alteration of mica by replacement of K with Ca, Mg, and H ions, as was shown to take place when biotite is leached with a $MgCl_2$ solution (2). A more detailed discussion of the nature of hydrous mica and its relation to vermiculite and montmorillonite will be presented elsewhere.

Discussion

The conversion of a contracted lattice, like biotite, to an expanded lattice, like vermiculite, by merely exchanging the K ions with Mg ions (2) warrant a closer examination of the relation between expansion and the exchangeable ions.

It is well known that the interlayer K ions in the contracted mica lattice occupy the cavities formed by the hexagonal rings of oxygens on the bases of the linked silica tetrahedra of opposite lattice layers (5). It is probable that in the contracted lattice found when the interlayer cation is K, NH_4, Rb, or Cs and when Li, Na, Mg, Ca, and Ba vermiculite lattices are dehydrated, the interlayer cations also occupy similar positions.

Calculations show that the radius of a sphere that just equals the size of the cavity within the hexagonal network of O ions is 1.35 Å. Comparison of the radius of a sphere of this size and the radii of the interlayer ions indicates that the radius of the interlayer cations may have an important bearing on the expansion of the lattice. In vermiculite when the interlayer positions are occupied by monovalent ions having a radius much smaller than 1.35 Å, namely, Li and Na ions, the lattice is expanded equivalent to a thickness of a monomolecular layer of water, whereas those ions with either an equal or a larger radius, namely, K, NH_4, Rb, and Cs ions are associated with an essentially contracted lattice. Furthermore when the radius of the exchangeable ion is at a minimum, for example the H ion, the expansion of the lattice becomes still greater and is equivalent to a thickness of two monomolecular layers of water. Among the divalent ions, those having radii substantially less than 1.35 Å cause expansion of the lattices about twice that of the Ba ion which is equal to 1.35 Å.

A comparison of the expansion of the crystal lattices with interlayer ions of equal size but different in charge, namely, monovalent versus divalent ions, suggests that the charge of the ions is an important factor in lattice expansion. Thus Mg or Ca ions cause about twice the expansion resulting from Li or Na ions. On the other hand, Ba ions bring about expansion equivalent to a monomolecular layer of water, whereas the K ions cause no expansion whatever.

Another factor appears when a comparison is made between the total interlayer charge and the degree of expansion of margarite, vermiculite, and montmorillonite. The essential difference between these three minerals, of significance in the present discussion, lies in the total negative charge neutralized by the interlayer cations. This charge per half unit cell is approximately 2 in margarite, from 1.0 to 0.7 in vermiculite, and from 0.5 to 0.3 in montmorillonite (14). Thus when the total charge is at a minimum, as in Ca montmorillonite, the lattice may expand to a width greater than that of two monomolecular layers of water; when the charge is at a maximum, as in margarite, there is no expansion at all (5); but when the charge is of an intermediate value, as in Ca vermiculite, expansion is limited to a thickness equivalent to two monomolecular layers of water.

A comparison of expansion of vermiculite and montmorillonite in glycerol and in water in terms of number of monomolecular layers of the interlayered substance rather than in terms of Å units reveals a very significant difference in expansion. Thus the total interlayer expansion of the vermiculites when immeresd in water is equal to two monomolecular layers but when immersed in glycerol it is only equal to one layer. Similarly the total interlayer expansion of montmorillonites when immersed in water is equal to four or more monomolecular layers but when immersed in glycerol it is only equal to two layers. Therefore it may be concluded that expansion of these minerals in glycerol is considerably less than in water and that the similarity in expansion when expressed in Å is the result of differences in size or shape of the molecules of the interlayered substance.*

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* An extensive study of the effect of the nature of the interlayered substance on expansion of vermiculites and montmorillonites is nearly complete. The results indicate that the dipole moment, the dielectric constant, and in some instances the form of the molecules are all factors in the expansion of dehydrated and contracted vermiculites and montmorillonites.

Therefore, it seems probable that: (1) the size of the interlayered ion, (2) the charge of the interlayered ion, (3) the total excess negative charge per unit cell of the mineral, and (4) the nature of the interlayered substance, are all factors in expansion.

In a separate discussion it will be suggested that the cause for these factors in expansion is related to their effect upon the interlayer attractive forces which hold the mica sheets together.

SUMMARY

1. The interlayer expansion was determined for air dry, water saturated, or glycerol saturated, vermiculite and montmorillonite containing any one of the following interlayer cations: Mg, Ca, Ba, H, Li, Na, K, NH_4, Rb, or Cs.

2. The interlayer expansion of air dry vermiculite and montmorillonite saturated with Mg, Ca, or H ions is equal to two monomolecular layers of water, but those saturated with Ba, Li, or Na ions are equal to only one molecular layer of water.

3. Air dry vermiculite saturated with K, NH_4, Rb, or Cs ions shows no expansion in terms of a monomolecular layer of water, whereas montmorillonite saturated with the respective ions shows an expansion of one molecular layer of water.

4. When immersed in water vermiculite saturated with Mg, Ca, Ba, H, Li, or Na shows a total expansion of only two monomolecular layers of water, but montmorillonite saturated with the respective ions shows an expansion of four or more monomolecular layers of water.

5. When immersed in water, K, NH_4, Rb, or Cs vermiculite shows no expansion, but the respective montmorillonite shows an expansion of one molecular layer of water.

6. When immersed in hot anhydrous glycerol, Mg, Ca, Ba, H, Li, or Na vermiculite shows an expansion of one layer of glycerol molecules whereas the respective montmorillonite shows an expansion of two layers of glycerol molecules. This results in an interlayer spacing of 14.3 Å for vermiculite and 17.7 Å for montmorillonite.

7. When immersed in hot anhydrous glycerol, K, Rb, or Cs vermiculite remains contracted, but the respective montmorillonite expands to a thickness of one molecular layer of glycerol.

8. NH_4 vermiculite remains contracted in hot glycerol but NH_4 montmorillonites expand to a thickness of either one or two layers of glycerol molecules.

9. Finely ground paragonite hydrates and expands.

10. The nature of hydrous mica clay minerals is discussed briefly.

11. Interlayer expansion of the mica type of a crystal structure was found to depend upon the size, the valency, and the total number of the interlayer cations as well as the nature of the expanding substance.

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NOMOGRAPHS FOR TRICLINIC CELL COMPUTATIONS

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Some of the equations used in computing the constants of triclinic cells, although rather involved, can be readily handled by nomographs. Given these nomographs one has a method of solving many triclinic cell problems very quickly with an accuracy of a fraction of a degree. Two types of problem are common. In the first type we are given the cell edge ratios and angles and wish to make a gnomonic projection to help in identifying a crystal that has been measured on an optical goniometer. In the second type of problem we are given certain atomic plane spacings and wish to compute the cell edges and angles and perhaps also to compute other atomic plane spacings.

We will consider the first type of problem first. As an example let us assume a crystal for which $a:b:c=0.900:1:0.800$, $\alpha=100°$, $\beta=95°$, $\gamma=105°$. The reciprocal angle chart, Fig. 1 gives us the reciprocal cell angles: α^*, β^*, γ^*. To find α^* we join the point β, γ of the grid to the point α of the linear α scale. This line crosses the linear scale α^* at a point that indicates the value of α^*. In our example the point 95°, 105° is found (on the right side of the grid because 95 and 105 are "like" in being greater than a right angle) and when joined to the point $\alpha=100°$ we find the line crosses the α^* scale at $\alpha^*=78.2°$. The point 105°, 100° of the grid joined to 95° on the center scale gives $\beta^*=82.0$ on the lower scale. Similarly we find $\gamma^*=73.8°$.

With a slide rule, we now evaluate the four quantities

$$V_1 = -\sin\alpha^*\cos\beta = 0.0853$$

$$V_2 = \sin\alpha^*\sin\beta = 0.975$$

$$A = \frac{\sin\alpha}{a\sin\beta} = 1.098$$

$$C = \frac{\sin\gamma}{c\sin\beta} = 1.212$$

We can now easily write the matrix $\overline{\mathrm{m}}^{-1}$:

$$\overline{\mathrm{m}}^{-1} = \left\{\begin{matrix} A\sin\gamma^* & 0 & CV_1 \\ A\cos\gamma^* & 1 & C\cos\alpha^* \\ 0 & 0 & CV_2 \end{matrix}\right\} = \left\{\begin{matrix} 1.055 & 0 & .103 \\ .307 & 1 & .247 \\ 0 & 0 & 1.182 \end{matrix}\right\}.$$

To make a gnomonic projection, on, say the plane perpendicular to the c axis, we evaluate the product $\overline{\mathrm{m}}^{-1}\left\{\begin{matrix} h \\ k \\ l \end{matrix}\right\}$ for a number of planes (hkl). On coordinate paper we plot x/z, y/z for each point. This is the gnomonic projection.

For example, for the plane (112) of the crystal we have been considering:

$$\begin{Bmatrix} 1.055 & 0 & .103 \\ .307 & 1 & .247 \\ 0 & 0 & 1.182 \end{Bmatrix} \begin{matrix}(1)\\(1)\\(2)\end{matrix} = \begin{Bmatrix} 1.055\times 1 + 0\times 1 + .103\times 2 \\ .307\times 1 + 1\times 1 + .247\times 2 \\ 0\times 1 \qquad + 0\times 1 + 1.182\times 2 \end{Bmatrix} = \begin{Bmatrix} 1.261 \\ 1.801 \\ 2.364 \end{Bmatrix}.$$

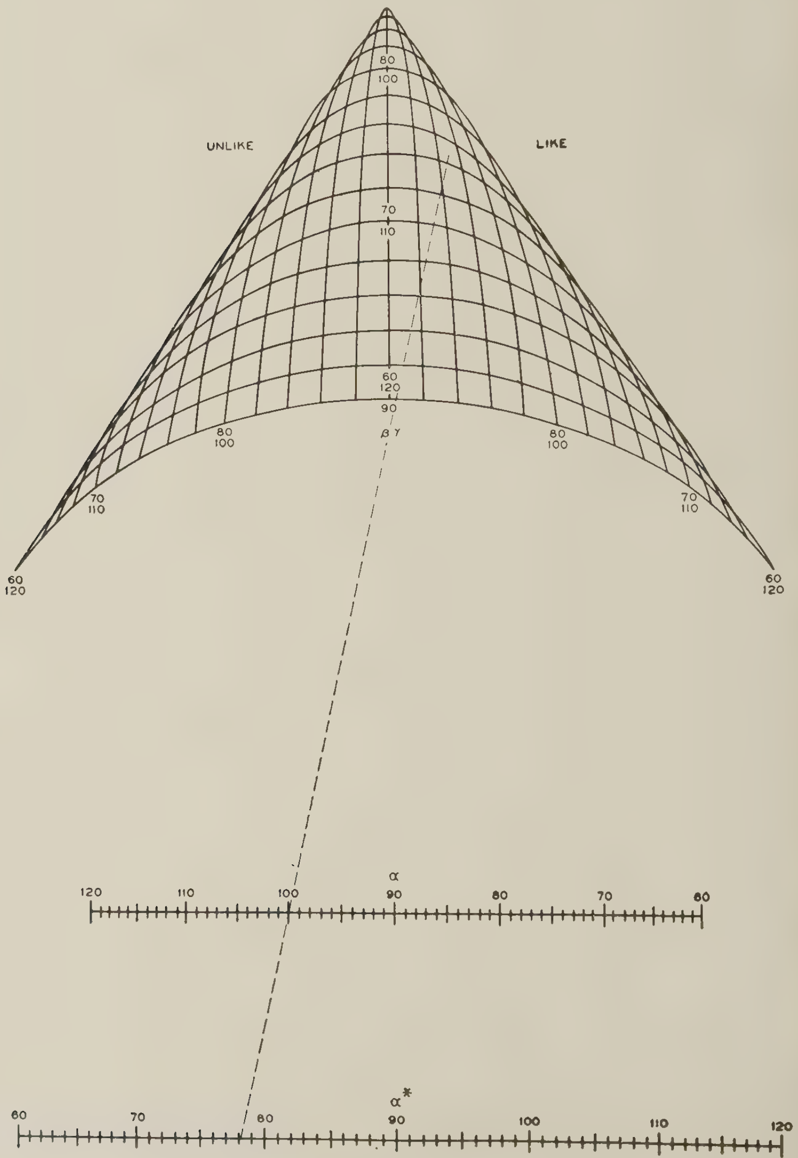

Fig. 1

The point $x=1.261/1.182=1.067$. $y=1.801/2.364=.762$ is the gnomonic projection of the plane (112).

If now, by x-rays we determine the value of d_{010} we can divide all terms of $\overline{m}^{-1}$ by d_{010} to obtain the matrix $\overline{M}^{-1}$. This matrix then gives all atomic spacings readily. We will illustrate the use of charts with this $\overline{M}^{-1}$ matrix by means of the next example, the second type of problem.

Let us assume that the measured reciprocal cell constants of a crystal are: $A_0=1/d_{100}=0.170$, $B_0=1/d_{010}=0.150$, $C_0=1/d_{001}=0.200$, $D_{01\bar{1}}==1/d_{01\bar{1}}=0.220$, $D_{10\bar{1}}=1/d_{10\bar{1}}=.260$ and $D_{110}=1/d_{110}=.204$. We know that the reciprocal cell angle α^* is determined by the ratios of B_0, C_0 and D_{011} (or $D_{01\bar{1}}$). We write these as a ratio in this order, then divide all three terms by the larger of the first two as $B_0:C_0:D_{01\bar{1}}=0.150:0.200:0.220$ $=0.75:1:1.10=e:l:g$ (or $l:e:g$ if the first number had been the larger of the first two). We then turn to the parallelogram chart, Fig. 2, and run

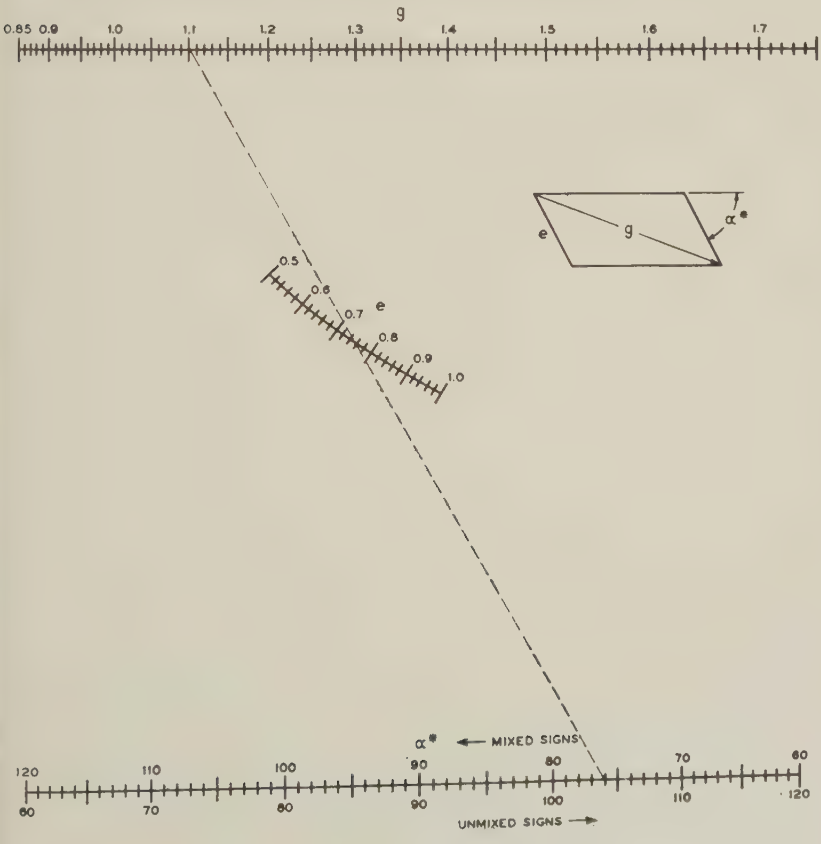

FIG. 2

a line through $g=1.10$ and $e=0.75$, then observe that this line cuts the α^* scale at 76.4°. (We read the upper numbers because $d_{01\bar{1}}$ has mixed signs.) Similarly $C_0:A_0:D_{101}=0.200:0.170:0.260=1:0.85:1.30$ gives $\beta^*=88.9°$ and $A_0:B_0:D_{110}=0.170:0.150:0.204=1:0.882:1.20$ gives $\gamma^*=101.1°$, (the lower numbers are read because D_{110} has unmixed signs).

To find α, β, γ from the angles α^*, β^*, γ^*, that we have just determined we use the reciprocal angle chart, Fig. 1 as before, merely considering starred angles to be unstarred and vice versa. The chart gives $\alpha=104.1°$, $\beta=93.8°$, $\gamma=78.4°$. A slide rule now gives us

$$a_0 = \frac{1}{\mathrm{A}\sin\beta\sin\gamma^*} = \frac{1}{\mathrm{A}\sin\beta^*\sin\gamma}, \quad \text{also}$$

b_0 and c_0 from the similar equations derived by permuting the letters. These equations give for our example, $a_0=6.01$, $b_0=7.00$, $c_0=5.16$. We now compute

$$\begin{aligned} \mathrm{V}_1 &= -\sin\alpha^*\cos\beta \\ \mathrm{V}_2 &= \sin\alpha^*\sin\beta \end{aligned}$$

for our example, getting $\mathrm{V}_1=.0642$, $\mathrm{V}_2=.970$.

We may now form the matrix $\overline{M}^{-1}$:

$$\overline{\mathrm{M}}^{-1} = \left\{\begin{array}{ccc} \mathrm{A}_0\sin\gamma^* & 0 & \mathrm{C}_0\mathrm{V}_1 \\ \mathrm{A}_0\cos\gamma^* & \mathrm{B}_0 & \mathrm{C}_0\cos\alpha^* \\ 0 & 0 & \mathrm{C}_0\mathrm{V}_2 \end{array}\right\} = \left\{\begin{array}{ccc} .1668 & 0 & .0128 \\ -.0326 & .150 & .0470 \\ 0 & 0 & .1940 \end{array}\right\}.$$

We will illustrate the computation of a plane spacing for the plane (312).

$$\left\{\begin{array}{ccc} .1668 & 0 & .0128 \\ -.0326 & .150 & .0470 \\ 0 & 0 & .1940 \end{array}\right\} \begin{array}{c} (3) \\ (1) \\ (2) \end{array} = \left\{\begin{array}{c} .1668\times 3+0\times 1+.0128\times 2 \\ -.0326\times 3+.150\times 1+.0470\times 1 \\ 0\times 3+0\times 1+.1940\times 2 \end{array}\right\}$$

$$= \left\{\begin{array}{c} .5260 \\ .1462 \\ .3880 \end{array}\right\} = \left\{\begin{array}{c} D_1 \\ D_2 \\ D_3 \end{array}\right\}.$$

The square root of the sum of the squares of these numbers is $1/d_{312}$ but the nomograph, Fig. 3 gives this value, $D_{312}=1/D_{312}$ much more easily and gives the Bragg angle for copper Kα besides.

We locate two of the three numbers on scales D_1 and D_2 and make a mark where a line joining these points crosses the ungraduated line. This crossing point is then joined to the third D value located on scale D_3 by a line that crosses scale D_{hkl}. This last crossing point gives $1/d_{hkl}$ below the line and the Bragg angle for copper Kα above the line. In our example this last crossing point is at $D_{hkl}=0.668$, hence, $d_{312}=1.497$ and above the line we read $\theta=31.0$.

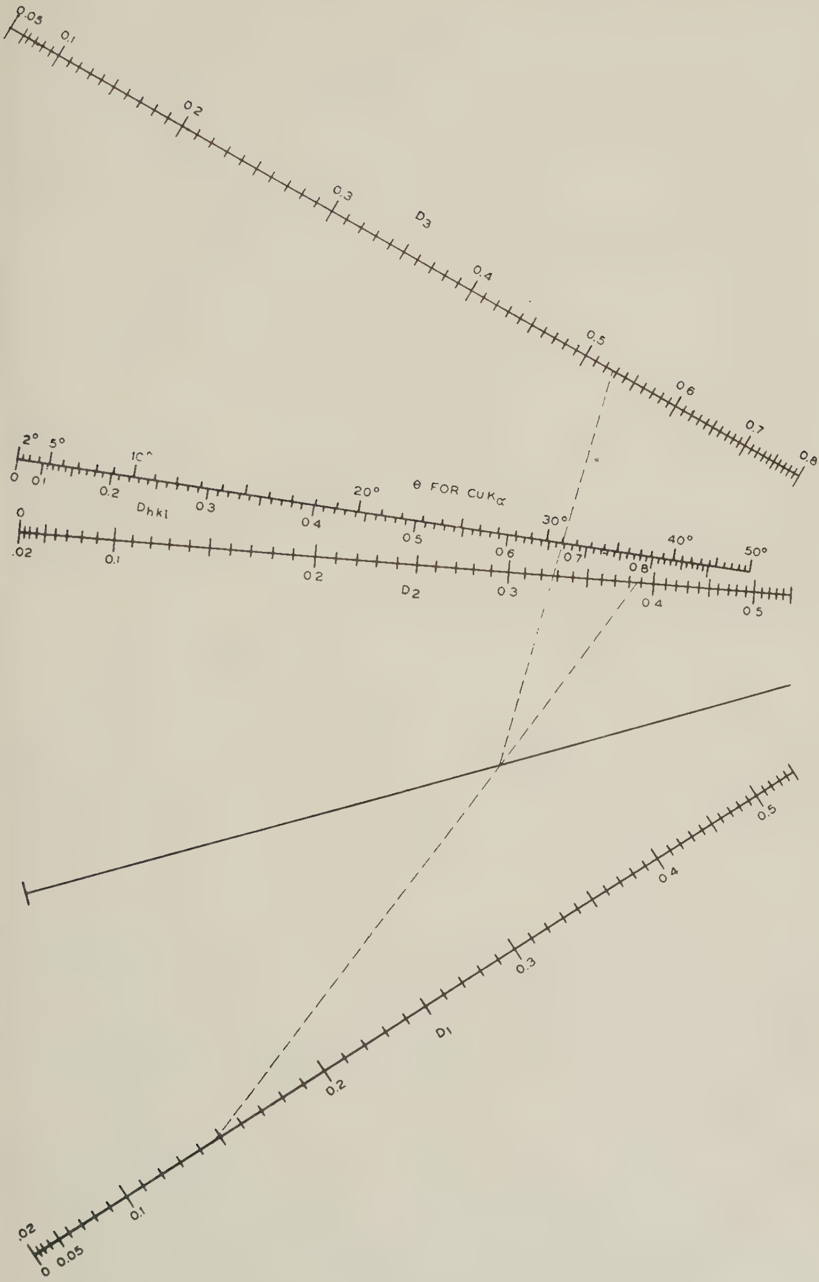
0.05
0.1
0.2
0.3
D3
0.4
0.5
0.6
0.7
0.8
2°
5°
10°
20°
θ FOR CuKα
30°
40°
50°
0
0.1
0.2
Dhkl
0.3
0.4
0.5
0.6
0.7
0.8
0
.02
0.1
0.2
D2
0.3
0.4
0.5
D1
.02
0
0.05
0.1
0.2
0.3
0.4
0.5

FIG. 3

While the charts published with this paper will be adequate for most purposes some workers may wish to prepare their own charts. The charts here presented may be photostated and hence enlarged to any convenient scale. Photographic distortion is immaterial as long as all straight lines are reproduced as straight. For those who wish to replot the charts we give the formulae

The Reciprocal Angle Chart.

The Grid: $x = \dfrac{\cos\beta\cos\gamma}{5/4 - \sin\beta\sin\gamma}$, $y = \dfrac{1}{5/4 - \sin\beta\sin\gamma}$

The α Scale: $x = 0.8\cos\alpha$, $y = .3$

The α^* Scale: $x = \cos\alpha^*$, $y = 0$

The Parallelogram Chart.

g scale: $x = 4/9g^2 - 5/6$, $y = 1$

e scale: $x = \dfrac{e^2 - 7/8}{2e + 9/4}$, $y = \dfrac{9/4}{2e + 9/4}$

α^* scale: $x = -\cos\gamma^*$, $y = 0$

The D Chart.

D_1 scale: $x = \dfrac{D_1^2}{1 + 10D_1^2}$, $y = \dfrac{-8.5}{1 + 10D_1^2}$

D_2 scale: $x = \dfrac{D_2^2}{1 + 10D_2^2}$, $y = \dfrac{1.5}{1 + 10D_2^2}$

D_3 scale: $x = \dfrac{D_3^2}{2 + 10D_3^2}$, $y = \dfrac{17}{2 + 10D_3^2}$

D_{hkl} scale: $x = \dfrac{D^2}{4 + 10D^2}$, $y = \dfrac{10}{4 + 10D^2}$

θ scale: $x = \dfrac{(2/\lambda)^2\sin^2\theta}{4 + 10(2/\lambda)^2\sin^2\theta}$.

In plotting these we do not need to plot x and y to the same scale. The derivation of these formulae will not be given here; it is a straightforward application of the methods given by the author in a recent paper—"A simple procedure for the making of alinement charts," *Journal of Applied Physics*, 83–86 (January 1948).

STUDIES OF URANIUM MINERALS (I):

PARSONSITE AND RANDITE*

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ABSTRACT

Parsonsite, $Pb_2(UO_2)(PO_4)_2 \cdot 2H_2O$, hitherto known only from the Belgian Congo, is described from the Ruggles pegmatite, New Hampshire, and from Wölsendorf, Bavaria. The New Hampshire material occurs as crusts of microscopic monoclinic crystals. The lath-like crystals are elongated on [001] and flattened on {010} with {001} as the terminal face; $\beta=99°$. Color citron-yellow; hardness $2\frac{1}{2}$ to 3; specific gravity 5.37; $c_0=6.8$Å. Analysis gave: PbO 47.43, UO_3 34.68, P_2O_5 14.46, H_2O 3.43; total 100.00 after deduction of 5.64 per cent quartz. The indices of refraction vary, apparently with the water content; most crystals have $nX \sim 1.870$, $nZ \sim 1.890$, $X \wedge c \sim 19°$. Parsonsite also occurs at Wölsendorf as microscopic yellow crystals identical in habit with the Belgian Congo and New Hampshire material. Biaxial negative, with indices of refraction, somewhat variable, of $nX \sim 1.795$, $nZ \sim 1.815$ and $Z \wedge c \sim 14°$. The x-ray powder spacing data are given.

Randite, originally described from Philadelphia, Pennsylvania, by Koenig in 1878 as a carbonate of calcium and uranium is discredited as a mixture consisting principally of calcite and beta-uranotile with some tyuyamunite.

PARSONSITE

In January of 1942, Mr. Philip C. Foster of the Boston Mineral Club submitted a small specimen from the Ruggles pegmatite near Grafton Center, New Hampshire, for identification. A preliminary examination at that time by the late Harry Berman showed the substance to be a uranium mineral but further work was deferred for lack of sufficient material. In January of 1946 a number of large specimens of the same mineral were obtained from a new find made by Mr. H. L. Smith of the Whitehall Company of Keene, New Hampshire, operators of the Ruggles mine. A quantitative analysis by F. A. Gonyer of this material, cited in Table 1, proved it to have the composition $Pb_2(UO_2)(PO_4)_2 \cdot 2H_2O$. This composition is identical with that attributed to parsonsite aside from a slightly higher content of water. The x-ray powder pattern, cited in Table 2, was found to be identical with a standard pattern of parsonsite from the files of the U.S. Geological Survey.

Parsonsite was originally described by Schoep from Kasola in the Belgian Congo, where it was found associated with torbernite, kasolite and dewindtite. The mineral occurred as aggregates of microscopic lath-like crystals with the crystal habit and optical orientation shown in Fig. 1. The observed morphological development and the inclined extinction indicates that the crystal symmetry is either monoclinic or triclinic. The plane of flattening was taken by Schoep and by Buttgenbach as {010} and the bounding forms, seen only under the microscope, were made {100}, {101} and {001}. The color of the aggregates is chocolate-brown, due to numerous included particles of foreign material, but the mineral itself is colorless to pale rose. Schoep assigned the formula $Pb_2U(PO_4)_2 \cdot H_2O$ to the mineral from the chemical analysis cited in Table 1. The analysis apparently was made on very impure material.

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* Contribution from the Department of Mineralogy and Petrography, Harvard University, No. 311.

TABLE 1. CHEMICAL ANALYSES OF PARSONSITE

	1	2	3	4
PbO	50.01	49.03	47.43	44.71
UO_3	32.06	31.42	34.68	29.67
P_2O_5	15.91	15.59	14.46	15.08
H_2O	2.02	3.96	3.43	1.56
Rem.				8.25
Total	100.00	100.00	100.00	99.27
S.G.			5.37	6.23

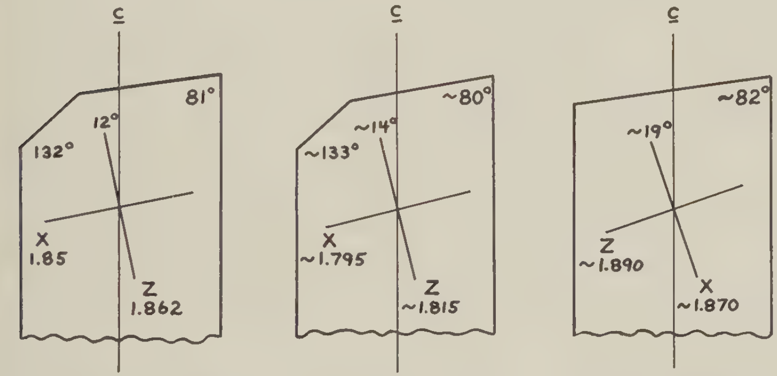
1. $Pb_2(UO_2)(PO_4)_2 \cdot H_2O$.
2. $Pb_2(UO_2)(PO_4)_2 \cdot 2H_2O$.
3. Parsonsite. Ruggles mine, New Hampshire. F. A. Gonyer analyst, April, 1947. Recalculated to 100 after deduction of 5.64 per cent insoluble.
4. Parsonsite. Kasola, Belgian Congo. Schoep analyst (1923). Rem. is CaO 0.63, CuO 0.25, Al_2O_3 1.23, TeO_3 3.01, MoO_3 0.43, CO_2 1.19; insol. 1.51. Original total given as 99.47. Analysis on impure material.

TABLE 2. X-RAY POWDER SPACING DATA FOR PARSONSITE

Copper radiation, nickel filter (in Å)

d	*I*	*d*	*I*	*d*	*I*
4.13	8	2.42	1	1.87	2
3.90	1	2.33	1	1.83	4
3.38	9	2.29	2	1.77	4
3.22	10	2.24	2	1.71	4
3.14	3	2.21	2	1.64	7
2.97	1	2.18	2	1.61	2
2.89	2	2.10	6	1.57	2
2.77	3	2.02	2	1.53	1
2.69	1	1.96	2	1.50	1
2.61	2	1.93	1	1.42	3
2.52	1	1.90	1	1.39	2
				1.36	3

Parsonsite from New Hampshire. Parsonsite occurs at the Ruggles pegmatite near Grafton Center, Grafton County, New Hampshire, as crusts of microscopic spicular or lath-like crystals. The mineral occurs sparingly along fracture surfaces in massive feldspar and quartz in the near vicinity of the uraninite and gummite masses for which the locality is known. The parsonsite is associated with autunite and phosphuranylite and all of these have ultimately been derived from the alteration of the primary uraninite. The color of the mineral is pale citron-yellow, and the luster is adamantine. The hardness is 2½ to 3, and the specific gravity, measured on a relatively compact aggregate, is 5.37. This value is much lower than the value 6.23 reported by Schoep for the material from the Belgian Congo. The latter value seems high for a substance of the present composition in comparison with the specific gravities of lead-uranium minerals such as dewindtite, renardite, and fourmarierite. It is not fluorescent in short- or long-wave ultraviolet light. Under the microscope, the crystals are seen to have a habit very similar to those from the Belgian Congo, although the optical properties are somewhat divergent therefrom. The crystals ordinarily are bounded only by {010}, {100} and {001}, with $\beta \sim 100°$; minute faces of {*hk*0} and {*hkl*} were noted in a few instances. The indices of refraction vary both within the same crystal and between different crystals. Most crystals have transparent and relatively perfect terminations and grade along the elongation toward the point of attachment into translucent material with much lower indices of refraction and with rough external faces. The terminal



1. Belgian Congo 2. Wölsendorf 3. New Hampshire

FIG. 1

parts of the majority of the crystals have the optical orientation shown in Fig. 1 with nX about 1.870 and nZ about 1.890. Other crystals have comparable birefringence but with lower values for nZ ranging down to about 1.86. A few crystals were noted with nX over 1.88. The indices of the Belgian Congo crystals were originally given by Schoep as over 1.99, with the optical orientation shown in Fig. 1, but Billiet later reported the values $nX=1.85$ and $nZ=1.862$. The optical orientation of most of the crystals from New Hampshire, as shown in Fig. 1, has the positions of X and Z opposite to those of the Belgian Congo material, but other crystals conformed to the latter orientation. The extinction angle between Z (or X) and c varied from about 6° to 23°, as measured on crystals clearly resting on $\{010\}$, and always was in the obtuse angle of β. The crystals are pale yellow in transmitted light and are not perceptibly pleochroic. No clear correlation could be found between the variation in extinction angle and either the optical orientation or the indices of refraction. The variation in the indices of refraction, found also in the parsonsite from Wölsendorf described beyond, presumably is connected with variation in the water content of the mineral at ordinary temperatures, as is characteristic of torbernite and some other hydrated oxysalts of uranium. The water content of the New Hampshire mineral as found by analysis is between $1H_2O$ and $2H_2O$ in the derived formula. The formula with $2H_2O$ is regarded as the more probable. The analysis was made by conventional procedures on a sample weighing only about 0.4 gram that contained about 6 per cent of admixed quartz and hence may be somewhat in error.

An effort was made to obtain crystallographic data by the x-ray rotation and Weissenberg methods, but the crystals were too small to yield satisfactory data. The identity period along the direction of elongation, or c-axis, was found to be 6.8 ± 0.1 Å. The Weissenberg resolution of the 0-layer about this axis was very faint, but the observed reflections were consistent with the plane symmetry C_{2l}. This fact and the inclined extinction against this axis indicate that the symmetry of the mineral is monoclinic with the elongation along c. The 0-layer photograph as projected gave a rectangular net with 10.21 Å and 6.77 Å as the dimensions of the smallest direct cell. One of these dimensions is b_0 or a submultiple thereof and the other is $a_0 \sin(\beta-90°)$ or a submultiple thereof, but their separate identity is not known. Efforts to obtain a 1-layer photograph about this axis or photographs about the b-axis were unsuccessful.

Parsonsite from Wölsendorf, Bavaria. A second new occurrence of parsonsite was found on a specimen in the Wölsendorf suite of the Harvard collection. This specimen comprised a mass of purplish black fluorite together with rusty-appearing quartz crystals and yellow plates of uranocircite. The parsonsite, identified by its x ray powder photograph, formed yellow, tufted aggregates of microscopic lath-like crystals deposited upon the uranocircite. The crystals were much smaller than those from New Hampshire and it did not prove possible to obtain physical or morphological data. Under the microscope the crystals had the habit shown in Fig. 1. The indices of refraction varied considerably but always were below those of the material from the Belgian Congo and from New Hampshire. Much of the material had the optical orientation shown in the drawing with nX about 1.795 and nZ about 1.815 and with $Z \wedge c$ mostly between 8° and 20° in the obtuse angle. A few crystals had nZ considerably below 1.81, with nX undetermined, and others had nZ about 1.825. Several crystals were noted with the reversed optical orientation previously found in the New Hampshire material. The extinction angle between Z (or X) and c ranged from a few degrees up to 36°. The crystals were colorless to faint yellow in transmitted light and did not show perceptible pleochroism. The optical sign was negative, and the dispersion strong.

The occurrence of parsonsite at Wölsendorf has been mentioned without description by Schoep and Scholz.

RANDITE

The name randite was given by Koenig in 1878 to a supposed hydrous carbonate of calcium and uranium found as canary-yellow coatings on granite pegmatite and hornblende gneiss at Frankford near Philadelphia, Pennsylvania. The original material came from Barber's quarry. Similar yellow coatings have been found on the Weissahickon gneiss at several nearby quarries associated with torbernite, autunite, and yellowish-green fluorescent hyalite. The original analysis, on 47 milligrams of material, was later shown by Rand to have been made on a mixture of calcite and an unidentified acicular mineral to which the name randite was then restricted. A partial analysis by Rand of the acicular crystals remaining after the calcite had been dissolved away by acetic acid gave CaO 16.71 and "U_2O_3" 46.71 per cent. A number of specimens of randite were available for examination including one specimen from the Academy of Natural Sciences of Philadelphia labelled as a gift from Mr. Rand. The writer is indebted to Mr. Samuel G. Gordon for opportunity to examine this specimen. Optical and x-ray study revealed that the yellow crusts were gross mixtures of calcite, kaolin, beta-urantile in minute acicular crystals, and tyuyamunite as thin dense crusts. The beta-urantile is predominant on the specimens and doubtless is the acicular mineral mentioned by Rand. Rand's partial analysis is considerably higher in CaO than either beta-uranotile or tyuyamunite, perhaps due to residual calcite. Whatever the explanation of this discrepancy, it appears that the name randite has been applied to a mixture of known species and hence is to be discredited.

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STUDIES OF URANIUM MINERALS (II): LIEBIGITE AND URANOTHALLITE*

HOWARD T. EVANS, JR. AND CLIFFORD FRONDEL, *Massachusetts Institute of Technology and Harvard University, Cambridge, Mass.*

ABSTRACT

Liebigite, $Ca_2U(CO_3)_4 \cdot 10H_2O$, from Joachimsthal, Bohemia, has a_0 16.71 Å, b_0 17.55, c_0 13.79; $a_0:b_0:c_0=0.952:1:0.786$ ($a:b:c=0.9539:1:0.7826$, morphology, Brezina, 1890). Space group *Bbam* or *Bba*. Cell contents $8[Ca_2U(CO_3)_4 \cdot 10H_2O]$. Specific gravity 2.41 (measured), 2.43 (calculated). The x-ray powder data, morphological data and new measurements of the optical constants are tabulated. Optically biaxial positive with $r>v$ and $2V \sim 40°$; $nX=1.497$, colorless; $nY=1.502$, pale greenish yellow; $nZ=1.539$, pale greenish yellow, as an average of values varying slightly among 12 different specimens. Uranothallite is shown to be identical with liebigite. A new occurrence of liebigite at Wheal Basset, Redruth, Cornwall, England, is noted and the occurrence at Schneeberg, Saxony, is verified.

Liebigite was described in 1848 by the American chemist J. Lawrence Smith, at that time geologist to the Sultan of Turkey, Abdul Medjid. The mineral occurred as an alteration product of uraninite and was associated with chalcopyrite, gypsum, and a supposed new sulfate of uranium and calcium for which the name medjidite was proposed. The locality was given only as in the neighborhood of Adrianople, Turkey. The liebigite was very briefly described as forming mammillary concretions, apple-green in color, with an apparent cleavage in one direction and a hardness of 2–2½. Two analyses made on samples weighing 85 and 65 mg. indicated the substance to be a hydrated carbonate of calcium and uranium. The percentage weights reported, however, differ considerably from the values required by the formula $Ca_2U(CO_3)_4 \cdot 10H_2O$ established by later analyses on uranothallite—a species here shown to be identical with liebigite.

In 1917 E. S. Larsen showed that the optical properties of material labelled liebigite from Schneeberg, Saxony, were identical with those of the well-defined mineral uranothallite, and on these grounds urged the identity of the two species. Liebigite had not been earlier described or analyzed from Schneeberg, however, and while there is no question of the identity of Larsen's particular material with uranothallite, there is of course no certainty that his mineral is identical with the original liebigite of Smith. The writers were privileged to borrow an authentic specimen of liebigite and medjidite from Adrianople in the collection of the

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* Contribution from the Department of Mineralogy and Petrography, Harvard University, No. 312.

American Museum of Natural History through the courtesy of Dr. F. H. Pough. This material (no. 16847) comprised two small pieces of botryoidal uraninite (pitchblende) with thin crusts of an apple-green cleavable mineral. Qualitative tests proved this to be a hydrated carbonate of calcium and uranium, and the optical properties and x-ray powder diffraction pattern were identical with those of crystals of uranothallite from Joachimsthal, Bohemia. The identity of liebigite and uranothallite thus appears to be certain. The name liebigite has priority of five years over the *kalk-uran-carbonate* of Vogl from Joachimsthal to which the names flutherite and uranothallite were later applied by Weisbach in 1875 and Schrauf in 1882, respectively.

TABLE 1. OPTICAL DATA FOR LIEBIGITE

Biaxial positive (+). Dispersion $r>v$, perceptible to moderate. Pleochroism: X nearly colorless, Y pale greenish yellow, Z pale greenish yellow

Locality	nX	nY	nZ	$2V$	Source
Joachimsthal	1.494	1.498	1.538	42°	New data
Joachimsthal	1.494	1.501	1.541	40°	New data
Joachimsthal	1.496	1.502	1.541	40°	New data
Joachimsthal	1.498	1.502	1.535	41°	Larsen, 1921
Joachimsthal	1.499	1.501	1.540	small	Larsen, 1921
Joachimsthal	1.500	1.503	1.539	42°	Larsen, 1921
Joachimsthal		1.505		small	Larsen, 1921
Schneeberg	1.499	1.503	1.545	variable, mostly 40°	New data
Schneeberg	1.501	1.503	1.537	37°	Larsen, 1921
Schneeberg		1.505		small	Larsen, 1921
Wheal Basset	1.497	1.503	1.538	15°	New data
Adrianople	1.497	1.503	1.542	40°	New data
Average	1.497	1.502	1.539		

Liebigite was also identified in this study on five specimens from Joachimsthal, Bohemia (two of which were erroneously labelled as voglianite, a dubious hydrous sulfate of uranium), on one specimen from Schneeberg, Saxony (labelled liebigite), and on a specimen from the Wheal Basset, Redruth, Cornwall, England. The latter locality is new for the species. The Wheal Basset specimen was erroneously labelled bassetite. The optical data obtained on several of these specimens are given in Table 1 in comparison with the data of Larsen. The x-ray powder diffraction spacing data are given in Table 2.

TABLE 2. *X*-RAY POWDER SPACING DATA FOR LIEBIGITE

Copper radiation, nickel filter (in Å)

d	*I*	*d*	*I*	*d*	*I*
6.81	9	2.71	1	1.575	2
6.07	2	2.59	4	1.547	1
5.37	10	2.47	3	1.497	1
4.98	1	2.32	3	1.473	2
4.55	6	2.28	3	1.451	1
4.10	5	2.18	4	1.423	1
3.93	1	2.12	2	1.402	1
3.79	2	2.05	1	1.358	2
3.60	4	2.01	5	1.329	1
3.35	7	1.929	2	1.301	2
3.16	8	1.852	1	1.285	2
3.04	2	1.731	4	1.258	2
2.85	2	1.684	2	1.222	2
2.78	2	1.631	1	1.182	1
				1.167	1

CRYSTALLOGRAPHY

Liebigite usually occurs as granular or scaly aggregates and as thin crusts or films. The films often occur as fillings of narrow cracks along the layers of mammillary masses of pitchblende that are undergoing alteration. The material from Adrianople is of this nature and doubtless gave rise to Smith's description of the mineral as forming mammillary concretions. Botryoidal aggregates are rarely observed. Crystals of liebigite are uncommon and generally are indistinct with rounded edges and convex or vicinal faces. Material suitable for single-crystal *x*-ray work was obtained from a specimen from Joachimsthal, Bohemia, in the Harvard collection (no. 84600). The following data were obtained by the precession method using molybdenum radiation.

a_0 16.71 Å	Space group *Bbam* or *Bba*
b_0 17.55	$a_0:b_0:c_0=0.952:1:0.786$ (*x*-ray cell)
c_0 13.79	$a:b:c=0.9539:1:0.7826$ (morphology, Brezina).

The *x*-ray cell here found coincides with the cell selected by Brezina in his morphological study of crystals from Joachimsthal. The agreement in axial ratio of the *x*-ray and morphological cells is satisfactory, especially in view of the poor surface quality of the measured crystals. An angle table using the elements of the *x*-ray cell is given in Table 3. The space group must be given as either *Bbam* or *Bba* for lack of certain knowledge of the crystal class. The small size and fragility of the

Table 3. Angle Table for Liebigite

Orthorhombic; probably holohedral
$a:b:c = 0.952:1:0.786$; $p_0:q_0:r_0 = 0.826:0.786:1$
$q_1:r_1:p_1 = 0.952:1.211:1$; $r_2:p_2:q_2 = 1.272:1.050:1$

	ϕ	$\rho = C$	ϕ_1	$\rho_1 = A$	ϕ_2	$\rho_2 = B$
c 001	—	0°00′	0°00′	90°00′	90°00′	90°00′
b 010	0°00′	90 00	90 00	90 00	0 00	0 00
a 100	90 00	90 00	—	0 00	0 00	90 00
n 230	35 01	90 00	90 00	54 59	0 00	35 01
m 110	46 25½	90 00	90 00	43 34½	0 00	46 25½
o 210	64 33½	90 00	90 00	25 26½	0 00	64 33½
d 011	0 00	38 10	38 10	90 00	90 00	51 50
p 111	46 25½	48 45	38 10	57 00	50 26½	58 47
r 121	27 43	60 37	57 32½	66 05½	50 26½	39 31½
q 141	14 02	73 38	73 09½	76 32½	50 26½	21 26
t 311	72 24	68 57½	38 10	27 10½	21 58½	73 36½

Doubtful: *x* 787 *u* 343 *s* 232 *y* 8.15.8.

available crystals precluded making a test for piezoelectricity. The specific gravity, given as 2.14–2.15 by Brezina, was redetermined on 12 mg. of powder on the microbalance as 2.41. The five reported chemical analyses of uranothallite = liebigite from Joachimsthal (cited by Dana and by Hintze) indicate the formula of the mineral to be $Ca_2U(CO_3)_4 \cdot 10H_2O$. The calculated specific gravity for the unit cell contents $8[Ca_2U(CO_3)_4 \cdot 10H_2O]$ is 2.43. There is a distinct cleavage on {100}. Liebigite fluoresces a rather bright green in both long-wave and short-wave ultraviolet light.

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PRESENTATION OF THE ROEBLING MEDAL OF THE MINERALOGICAL SOCIETY OF AMERICA TO HERBERT E. MERWIN

Norman L. Bowen, *Geophysical Laboratory, Washington D. C.*

Mr. President, Fellows and members of the Mineralogical Society of America, and guests:

Our Roebling laureate was born at Newton, Kansas. He must have been a very precocious and determined little chap; at least the evidence seems to indicate that, at the age of two, he read Horace Greeley's admonition, "Go West, young man", and accordingly went East, taking his parents with him and settling in New York state. He grew to manhood in the Catskills where interesting geologic surroundings made their impress on an enquiring mind. After several years of teaching in the public schools and on the faculty of the Normal School at Oneonta, he was claimed by Harvard for undergraduate studies and subsequently for graduate studies. There he fell under the influence of William Morris Davis and his first published paper was on Vermont shore lines, but from assistant in physiography he passed to teaching fellow in mineralogy and petrography and in 1909 he published his first paper in mineralogy, a joint paper with Professor Charles Palache on the then new species, alamosite. His early experience in petrography was gained as a member of that unusual group of young men who worked with Professor John E. Wolff in the Crazy Mountains.

It was during this period that he acquired an absorbing interest in light, first manifested in his development of a compound light filter for use in examining the characteristic flames of the alkali metals, later to evolve into his life's work in crystal optics.

With this diverse background Merwin came to the Geophysical Laboratory in 1909 where he has remained ever since. He has been a participant in almost every aspect of the laboratory's investigations in these four decades. In studies of phase equilibrium the identification of phases by optical methods is not only an indispensable aid but a guide and control. He has had a prominent part in the development and extension of these optical methods. Special immersion media of unusually high refraction and again others of low refraction have been prepared and described by him. In 1912 he published, jointly with Larsen, his dispersion method of measuring refractive indices of grains in immersion liquids, a method requiring interpolation between values obtained by matching at more or less remote wave-lengths. In 1917 he published tables facilitating accurate interpolation.

Herbert Eugene Merwin

Recipient of the Roebling Medal of the Mineralogical Society of America

Applying his various methods, he has probably determined the optical properties of more synthetic mineral compounds than any other individual. He has also done a great deal of work on opaque minerals in reflected light, his investigations furnishing the control in studies of the synthesis of sulfides of zinc, cadmium, mercury, iron and copper.

Merwin is a reluctant writer and most frugal of words when he does write. The results of most of his work appear as a paragraph or two in papers of which he is the junior author. Seldom did any of his contributions appear under his sole name. Nevertheless, in spite of his excessively modest and retiring nature, he long since came to be generally recognized as a leading authority on crystal optics. He was, for instance, chosen to write for International Critical Tables the section on Refractivity of Birefracting Crystals.

We, his immediate colleagues, have leaned heavily upon him and know him as an authority on a great many other subjects as well. Whenever two or three of us were gathered together and fell to discussing some problem, if we reached an impasse one was sure to suggest "Ask Merwin." Usually our query had fruitful results.

Merwin is now on the retirement list, but fortunately for us he is still at work every day as usual, and we can still ask Merwin.

Mr. President, it is an honor and a pleasure to present Herbert Eugene Merwin, a past president of the Society, for the Roebling Medal.

ACCEPTANCE OF THE ROEBLING MEDAL OF THE MINERALOGICAL SOCIETY OF AMERICA

HERBERT E. MERWIN, *Geophysical Laboratory, Washington, D. C.*

As a boy in a dairying community, I learned a trade, that of making small tight barrels and tubs for storing butter in brine. I became greatly impressed by the special tools used and by the many particular properties required of wood for staves and for hoops. Much of my spare time was spent in reading, experimenting and botanizing. In the summer our shop doors were open, and men vacationing from the city came in. One such visitor who came in a second time brought to me a newspaper clipping telling of argon, the atmospheric gas just discovered, and of the formation of magnesium nitride in purifying it. I hadn't interpreted my reading as allowing such a compound, and the inadequacy of my reading became apparent. I gave up coopering and taught school for two years.

Then I entered a training school for teachers at Oneonta, New York, and found an enthusiastic science teacher, Howard Lyon, who had acquired for the school a well-filled cabinet of selected minerals. I was soon handling pitchblende and the spinthariscope. While Marconi was busy in England with wireless we were carrying a coherer beyond the local campus and catching signals from our physics laboratory. Not many months later I received week after week from a distant hospital reports of the progress of Lyon's recovery from an *x*-ray burn from one of our Crookes tubes. During the maple sugar seasons we made observations on sap pressure, and for two years I taught physical geography and biology.

In the excitement of university life I became acquainted with the goniometer, that instrument which marks the upbringing of the mineralogist and finally I was guided by Professors Charles Palache and John E. Wolff into Geophysical Laboratory of the Carnegie Institution of Washington, in the establishment of which Professor Wolff had been keenly interested a few years before.

During my forty years there my colleagues and I witnessed and took part in the development of methods of preparing and characterizing crystalline materials, and finding their physical and chemical relationships. Developments such as these were so varied and interrelated that cooperation was sought in research.

Thus, the world to me has been one of friendly associates, and now to the the Mineralogical Society of America I express my sincere gratitude for the Roebling Medal.

MEMORIAL OF FREDERICK NOEL ASHCROFT

F. A. BANNISTER, *British Museum (Natural History), London S.W.7*

Mr. Frederick Noel Ashcroft, one of the notable mineral collectors of his time and a scientific photographer of the first order died suddenly at his home in London on 4 April, 1949. His scientific interests combined with a flair for administration backed by financial acumen enabled him to render outstanding services as Treasurer both to the Geological Society of Great Britain 1929–1947 and to the Mineralogical Society 1924–1942.

Frederick Noel Ashcroft was born at Wavertree, Liverpool, on August 28th, 1878. He entered Rugby School in 1892 and proceeded to Magdalen College, Oxford in 1897 where he took a "first" in Chemistry Honours in 1901. In that year he also studied at Tübingen, Germany, and accompanied Sir Harold Hartley with whom he had worked at Oxford on a collecting trip to the Seiser Alpe, Bozen, Tirol.

Subsequently he carried out research work in organic chemistry at University College, London, and his first scientific publication in 1907 dealt with an attempt to synthesize a double pyrone ring compound. It was not long, however, before he was exhibiting to the Mineralogical Society zeolites from the neighbourhood of Belfast, and in 1914 he presented to the British Museum over 2,000 specimens of these minerals from all over the world and chiefly from the United States of America and Nova Scotia. His four cabinets of Irish zeolites were subsequently bequeathed to the British Museum.

From 1914–1918 opportunities to one so busily engaged in war work were restricted mainly to the English Lake District and to the mines of Derbyshire, West Cumberland and Wales. After the war he also made collecting trips to Wanlockhead and the Leadhills on which he was accompanied by Dr. Campbell Smith, now Keeper of Minerals in the British Museum. It was at this time that he first turned his attention, probably at the suggestion of Dr. L. J. Spencer, to Swiss minerals, and it is with these that Ashcroft's name will always be associated by mineralogists.

His Swiss collection is the finest ever made and comprises 6,625 specimens accompanied by photographs of the sites and meticulously written labels and catalogue entries. It is the most completely documented of any regional collection in the Mineral Department of the British Museum. "Die Mineralien der Schweizeralpen" by P. Niggli, J. Koenigsberger and R. L. Parker, to which Ashcroft contributed sixteen magnificent plates and a wealth of topographical detail, gives the key to his method of collecting minerals. Always he had in mind problems of mineral origin. His success in preserving the entire contents of each "Kluft" and his per-

Frederick Noel Ashcroft
1878–1949

severance in recording the mineral associations and their paragenesis will in due course bring their reward.

Ashcroft owed much to the encouragement of Sir Henry Miers in his undergraduate days, and it is natural that his interests should become embracing and his sympathies liberal ones. He became in 1920 one of the first Charter life Fellows of the Mineralogical Society of America and Managing Trustee of the Geologists' Association of Great Britain in 1925. As Treasurer and Trustee he secured the financial foothold of the Mineralogical Society and doubled its assets. He inaugurated a period of prosperity upon which depended the ever increasing size and quality of the Mineralogical Magazine and the Mineralogical Abstracts.

He leaves his wife, a daughter of the late John Conrad im Thurn, a son and two daughters. Fortunately, he was spared the knowledge that his young son, a brilliant mathematician and one of the outstanding men in the British Treasury, was to outlive him only by a few months.

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MEMORIAL OF EVEREND LESTER BRUCE

J. E. Hawley, *Queen's University, Kingston, Ontario.*

Dr. E. L. Bruce, Miller Memorial Research professor and head of the Department of Geology, Queen's University, Kingston, Ontario, died on October 5, 1949, following a heart attack suffered ten days previously while he was preparing for another University session. An earlier attack in March had enforced a long rest through the spring and early summer, after which he seemed to have made such a good recovery that he returned to his desk and became increasingly involved in his many duties.

Born in 1884 at Toledo, in the vicinity of Smith Falls, Ontario, Dr. Bruce received his undergraduate training at Queen's University obtaining the degrees of B.Sc. and B.A. in 1909 and 1911. His master's and doctorate work was carried out at Columbia University under Kemp and Berkey in 1912–1915, and a further year of post-graduate study was spent with Van Hise and Leith at Wisconsin.

Everend Lester Bruce
1884–1949

His early geological investigations (1912–1918) with the Ontario Department of Mines and Geological Survey of Canada led him to northern Ontario, Manitoba, and Saskatchewan where he developed a great love of the northland and an unfailing interest in the many problems of the pre-Cambrian. In 1919, Bruce came to Queen's University and succeeded N. L. Bowen as head of the department of Mineralogy where he systematized in typical fashion, and added substantially to the many mineral collections previously acquired by Professor William Nicol. Expanding his courses to the graduate level, he brought to his teaching of mineralogy and later, geology, a wealth of practical field experience which few instructors could equal.

Following the establishment of the Willet G. Miller Memorial Research Professorship in the Department of Geology, Bruce accepted this post in 1929 and began the development of graduate courses in pre-Cambrian, metamorphic, structural and economic geology, culminating in the publication of his book on Mineral Deposits of the Canadian Shield.

In 1944 he assumed the Chairmanship of the Committee on Graduate Studies for the entire university and in the same year succeeded M. B. Baker as head of the Department of Geology. There is no doubt that this heavy load of administrative duties contributed much to his untimely death.

Throughout his career many honours came to him. He was a fellow of the Geological Society of America, the Mineralogical Society of America, the Royal Society of Canada, and a member of the Geological Society of Edinburgh, the Geological Society of Finland, the American Institute of Mining and Metallurgical Engineers and the Canadian Institute of Mining and Metallurgy. His work was fittingly recognized on several occasions, when he was chosen as President of Section IV (Geological Sciences) of the Royal Society of Canada in 1933 and of the Geological Society of America in 1943. In 1948, he was elected Vice-President of the Society of Economic Geologists. Still other honours were to be his, had he lived.

Bruce's chief interest lay in the economic mineralogy and geology of metalliferous deposits throughout the pre-Cambrian as shown by his long record of active field work and resulting government reports and scientific papers on various mining areas. Field seasons not indicated in his bibliography were devoted to consulting geological work for various companies, in study of the pre-Cambrian of Finland in 1930 with Sederholm, or in attendance at International Geological Congresses held in Spain, Moscow, and England.

Outstanding among his explorations in virgin territories are those in which he laid an excellent foundation for the geology of the copper and gold areas of northern Manitoba and Saskatchewan, and the Red Lake gold area of Ontario. He also pioneered in the Little Long Lac area, Ontario, and contributed much in later years to the geology of the

Michipicoten district. Shorter periods were spent in the gold-copper belt of northwestern Quebec, the Rossland district in British Columbia, and more recent visits to the Northwest Territories and the Labrador iron deposits kept him in touch with still other phases of the mineral industry. It is little to be wondered at that in later years he came to be regarded as the dean of Canadian pre-Cambrian geologists.

Bruce's contributions to the science of Mineralogy consist chiefly of his keen and accurate observations of the occurrence of economic minerals and associated metamorphic alterations in the numerous deposits he examined. His was primarily "geological mineralogy" of the highest order, but he was always quick to recognize the unusual, as witness his several papers on odd or rare varieties of minerals encountered from time to time.

Of even more significance, however, are his contributions to the geological sciences as a teacher, both of numerous field assistants on surveys and of students in class room or laboratory. Here too he excelled as an administrator and director of research, and was tireless in his efforts to secure the best and most up-to-date equipment with which his students could work. Always a patient, modest, and kindly instructor, he had the faculty of drawing out the best, of leading without appearing to lead, and of imparting that personal interest which made all his students and colleagues his lasting friends.

His marriage to Mrs. H. C. Horwood of Ottawa in 1923 marked the beginning of the happiest part of his life, which continued unbroken till her death in 1943, after a lengthy illness. Her gay wit and charm and the kindly hospitality of both made their home a mecca for all geologically-minded as well as countless other friends. Their two sons, Douglas and Geoffrey survive.

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PROCEEDINGS OF THE THIRTIETH ANNUAL MEETING OF THE MINERALOGICAL SOCIETY OF AMERICA AT EL PASO, TEXAS

C. S. Hurlbut, Jr., *Secretary*

The thirtieth annual meeting of the Society, which was held on November 10–12, 1949 in El Paso, Texas, was attended by 134 members and fellows. Scientific sessions were held in the morning and afternoon of November 10th and the morning of November 12th, at which thirty-five papers were presented. Five additional papers were presented in the afternoon of November 11th at a well-attended Symposium on *Disorder in Minerals*.

The annual luncheon of the Society on November 11th was attended by 87 fellows, members and guests. Following the luncheon, Norman L. Bowen introduced the recipient of the Roebling Medal, Herbert E. Merwin of the Geophysical Laboratory, Washington, D. C. Dr. Merwin is the ninth recipient of the medal. Preceding the Symposium on *Disorder in Minerals*, the retiring President, John W. Gruner, addressed the Society on *An Attempt to Arrange Silicates in the Order of Their Free Energies.*

The Council at its meetings on November 9 and 10, 1949, in addition to transacting routine business of the Society, passed unanimously two motions that should be of interest to the general membership. The first was the new award of the Mineralogical Society of America for an outstanding contribution to mineralogy by a person of not more than thirty-five years of age. The specifications for this award were printed in the January-February 1950 issue of *The American Mineralogist.* The second was the election of

Charles Palache, Honorary President of the
Mineralogical Society of America.

Although the Constitution of the Society provides for election by the Council of honorary officers, Dr. Palache, Professor Emeritus of Mineralogy at Harvard University, is the second to be elected an honorary officer. The first Honorary President was Edward S. Dana, who served in that capacity from 1925 until his death in 1935. Dr. Palache was a charter fellow of the Society and served as its second President in 1921. He was the first recipient of the Roebling Medal, which was awarded to him in 1937.

At its meeting in 1948 the Council felt that the Constitution of the Society should be completely revised and brought up to date. Accordingly, a committee with Joseph Murdoch as Chairman was appointed by President Gruner to scrutinize the Constitution and bring before the Council at its next meeting any recommendations. The report of this committee was discussed at length, and several changes were accepted by the Council. The complete Constitution, incorporating these changes, will be printed in the July-August 1950 issue of *The American Mineralogist*, and the general membership at the next election will be asked to accept or reject these changes.

On the following pages are given the reports of the officers for the year ending October 31, 1949, as read before the Council at its meetings on November 9 and 10, 1949.

REPORT OF THE SECRETARY

To the Council of The Mineralogical Society of America:

Election of Officers and Fellows

Four hundred and thirty-one ballots were cast in the election of officers: 284 by members and 147 by fellows of the Society. The officers elected are:

President: George Tunell, University of California at Los Angeles, California.

Vice-President: Ralph E. Grim, Illinois Geological Survey, Urbana, Illinois.
Secretary: C. S. Hurlbut, Jr., Harvard University, Cambridge, Massachusetts.
Treasurer: Earl Ingerson, U. S. Geological Survey, Washington, D. C.
Editor: Walter F. Hunt, University of Michigan, Ann Arbor, Michigan.
Councilor (1950–53): E. F. Osborn, Pennsylvania State College, State College, Pennsylvania.

According to the provisions of the Constitution, the following have been elected to fellowship:

Charles Findlay Davidson, Geological Survey of Great Britain, London, England.
Gordon Leslie Davis, Geophysical Laboratory, Carnegie Institution, Washington, D. C.
Frank Ebbutt, Howe Sound Company, Toronto, Canada.
Austin Burton Edwards, University of Melbourne, Australia.
Albert E. J. Engel, California Institute of Technology, Pasadena, California.
George M. Furnival, Department of Mines and Natural Resources, Winnipeg, Manitoba.
Gonzalez-Bonorino, Felix, Direccion de Minas y Geologia, Buenos Aires, Argentina.
Wilfrid K. Gummer, Aluminum Laboratories Limited, Arvida, Quebec, Canada.
Maurice Hall Haycock, Bureau of Mines, Ottawa, Canada.
Elbridge Churchill Jacobs, University of Vermont, Burlington, Vermont.
Howard W. Jaffe, U. S. Bureau of Mines, College Park, Maryland.
Mackenzie L. Keith, Geophysical Laboratory, Carnegie Institution, Washington, D. C.
Paul D. Krynine, Pennsylvania State College, State College, Pennsylvania.
Kathleen Lonsdale, University College, London, England.
Charles E. Michener, The International Nickel Company of Canada, Ltd., Copper Cliff, Ontario, Canada.
Arie Poldervaart, Geological Survey, Bechuanaland Protectorate, South Africa.
A. Williams Postel, U. S. Geological Survey, Washington, D. C.
Rex Tregilgas Prider, University of Western Australia, Nedlands, Western Australia.
Thure Georg Sahama, University of Helsinki, Finland.
Ernest B. Sandell, University of Minnesota, Minneapolis, Minnesota.
Frederick Stewart Turneaure, University of Michigan, Ann Arbor, Michigan.
Francis J. Turner, University of California, Berkeley, California.
Samuel Zerfoss, Naval Research Laboratory, Washington, D. C.

CHANGES IN THE BY-LAWS

On the recent ballot, fellows and members were asked to vote on the following proposed change in Article II, Section 4 of the By-Laws:

From

A single prepayment of one hundred dollars ($100) shall be accepted as commutation for life for either fellows or members. In the case of fellows who are also fellows of the Geological Society of America, a single prepayment of fifty dollars ($50) shall be accepted as commutation for life.

To

A single prepayment of an amount equaling twenty times the annual dues of a fellow of the Society shall be accepted as commutation for life for either fellows or members.

This change was approved by a vote of 372 in the affirmative with 35 in the negative.

MEMBERSHIP STATISTICS

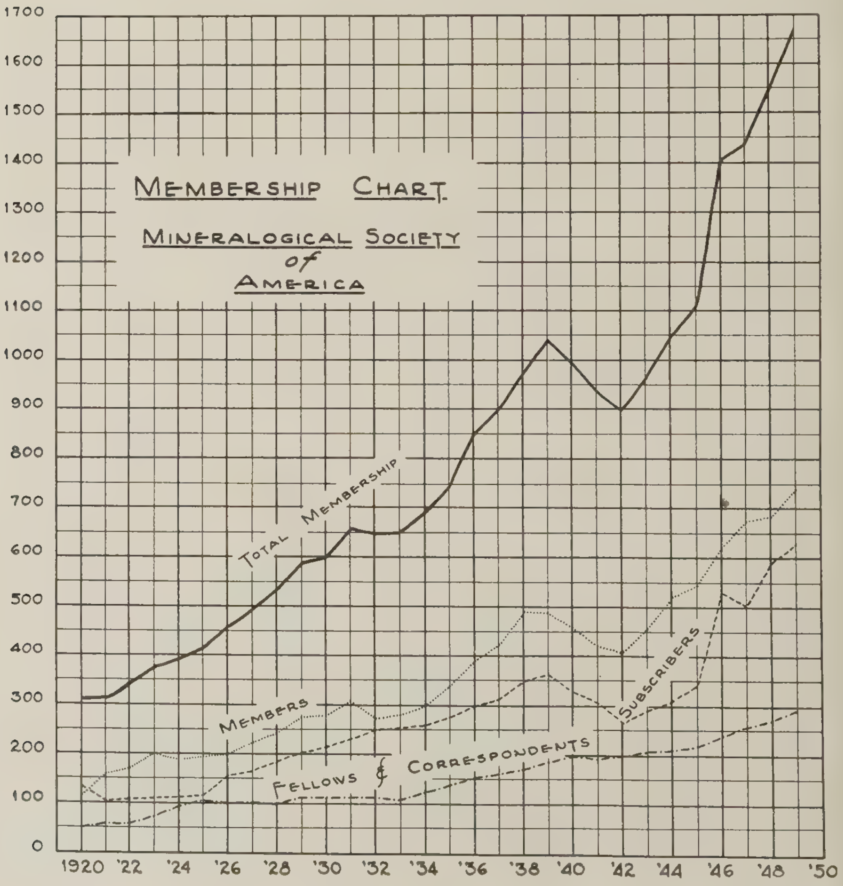
November 1, 1949

	1948	1949	*Gain*	*Loss*
Correspondents	5	5	0	0
Fellows	270	290	23	3
Members	686	746	182	122
Subscribers	592	630	91	53
	1553	1671	296	178

The above figures show a net gain of 20 fellows, 60 members and 38 subscribers. Considering the four groups together, there is a total gain of 118. The present total of members and subscribers is 1,671, which represents an increase of almost 8% since 1948.

The Society lost through death two fellows: F. N. Ashcroft, of London, England, and E. L. Bruce, of Kingston, Ontario, Canada.

Respectfully submitted,
C. S. Hurlbut, Jr., *Secretary*



REPORT OF THE EDITOR FOR 1949

To the Council of the Mineralogical Society of America:

In presenting the annual editorial report at this time, the same general policy will be followed as that pursued a year ago. With five issues published and distributed and only one still in press, sufficient data are now available to give a general survey of the year's activity. By the time the Editor's report appears in print (March-April issue) the missing data will be available and be incorporated, so that the then complete report can be compared on the same basis as the summaries given for last year and those of previous years.

From the standpoint of scholarly productivity I think the critical readers will agree that the Journal for 1949 has moved forward and has not merely marked time. From present indications the current volume will reach, or possibly exceed the 900 page mark, a substantial increase in size (approximately 100 pages) over that of a year ago. If this goal is realized it would represent the largest volume in the history of the Society with the single exception of the volume of 1937 that included the unusually large special Palache number.

In the main the policy of former years has been followed. Several excessively long manuscripts have been returned to authors for condensation and in several instances for reduction in the number of cuts originally submitted. This policy seems necessary not only to reduce as far as possible printing costs but also to speed up the production line by printing the largest number of manuscripts in the available space.

Unusually long manuscripts do impose at times special problems as they act as "road blocks" to rapid service by replacing several shorter articles. Also in some cases they represent highly specialized phases on the outer fringe of mineralogy or crystallography. In addition some are highly mathematical in character. In those cases I believe it can be truthfully stated that they are read in toto by very few readers, most of whom are quite content to merely glance at the abstract.

The question of how to reduce costs and expedite production is one not restricted to our Journal. The larger parent organization, the Geological Society of America with its large and efficient editorial staff and liberal financial resources, is facing the same problem. Permit me to quote one short paragraph from the Report of the Committee of Publications as recorded in the Interim Proceedings—Part 3, (Sept., 1949) page 32.

"The Committee also felt that economies could be effected at the source, and urged greater care in composition of papers by prospective authors. They recommended that the rising costs be impressed on Fellows, Members, and authors generally, and that authors be urged to condense all papers to a minimum. They further recommend that, in the editorial and publication procedure, short papers be given priority in publication procedure." This idea of giving shorter papers priority seems a splendid suggestion and one that I think we could follow with profit.

This year's volume included one colored folded insert of an article illustrating line and band spectra observed with a Bunsen spectroscope. It was felt that this article could be adequately illustrated only by employment of color. An innovation this year, sanctioned by the Council, which appears to have met with the hearty approval of our readers, was devoting the May-June number exclusively to *Contributions to Canadian Mineralogy.* In that issue the guest Editor, Dr. M. A. Peacock, assembled 15 interesting papers and assumed full responsibility in seeing these contributions through the press. I should like to express my appreciation to Professor Peacock for this timely and valuable assistance. Also recognition and gratitude is here expressed for the generous and continued assistance from the Geological Society of America in the form of financial support toward defraying a substantial portion of the ordinary publication costs and likewise for aid in reproducing and printing the colored insert.

With reference to the material now on hand and tentative commitments for 1950, mention might be made of the following:

(1) Presidential Address and 27 abstracts of papers given at the fourth annual spring meeting of the Crystallographic Society of America, held at Ann Arbor, Michigan, April 7–9, 1949.

(2) About 30 papers on general mineralogical subjects now on file, not including those in press to appear in the November-December issue.

(3) Contributions to Canadian Mineralogy will again very likely constitute a complete single issue and the May-June number may be used for this purpose.

(4) Arrangements are being made at Harvard for a special number and the September-October issue has tentatively been set aside for such an issue.

From this outline, even at this early date, it would seem that 1950 will be a very busy year. The size of the Journal will unquestionably equal and very likely exceed the current volume of 900 printed pages. Much depends upon the size of the special number as to the size of the volume for next year. Indications are, however, that no depression is in sight.

During the past year there has been no extremely heavy back log of accumulated manuscripts. The expansion in size of the individual issues thus far has been able to take care of the papers submitted. Ordinarily, the time interval between reception and appearance of

Table 1. Distribution of Subject Matter in Volume 34

Subjects	*Articles*	*Pages*	*Per Cent of Total*
Leading articles*			
Descriptive mineralogy	10		
Chemical mineralogy	22		
Structural crystallography	15		
Geometrical crystallography	6		
Petrography	3		
Optical crystallography	2		
Mineralography	1		
Memorials	5		
Miscellaneous	9		
	73	754	84
Shorter articles	25	54	16
Notes and news	20	9	
Proceedings of Societies	5	65	
Book reviews	10	11	
New mineral names	20	5	
Total entries	153	898	100
Illustrations	221		
Index, Title page, Table of contents		18	
Grand total		916	

* Leading articles average 10.3 printed pages each.

the printed article has been from six to eight months. In some few instances the delay has been slightly longer. This time interval may seem long to some authors who frequently ask for an early appearance, but to those who have had experience with other scientific publications the delay does not seem unreasonable for present conditions.

The current volume will contain 73 leading articles and 25 of a shorter character, totaling 98 published manuscripts for the year (last year it was 55 and 18, totalling 73). Twenty-eight contributions were received from contributors residing outside of the States, 21 of which were received from Canada. These contributions were received from 109 contributors associated with 52 different Universities, research bureaus, and technical laboratories.

Nine new mineral species were described in detail for the first time: frondelite, laubmannite, retgersite, rockbridgeite, scorzalite, sengierite, souzalite, torreyite and wolfeite.

The accompanying table 1 summarizes in detail the distribution of subject matter in volume 34.

Respectfully submitted
WALTER F. HUNT, *Editor*

(*Treasurer's report on next page.*)

REPORT OF THE TREASURER FOR 1949

To the Council of the Mineralogical Society of America:

Your Treasurer submits herewith his report for the fiscal year beginning November 1, 1948, and ending October 31, 1949.

Receipts

Cash on hand, November 1, 1948	$ 2,579.23
Dues and subscriptions	7,307.86
Sale of back numbers	1,237.83
Authors' charges on reprints	798.17
Income from endowment	3,761.85
Final payment of principal of Trenton Mortgage Co. stock	156.22
Geological Society of America aid in printing the Journal	6,328.75
Advertising	429.73
Sale of 20-volume Index	9.00
Sale of Index to volumes 21–30	124.19
Sale of securities	5,175.00
Securities called	1,050.00
	$28,957.83

Disbursements

Printing and distribution of the Journal (six issues)	$11,544.48
Printing and distribution of reprints	1,438.02
To the Editor, Secretary, and Treasurer	1,250.00
Postage and express	350.53
Clerical and secretarial assistance	514.25
Office equipment	150.55
Printing and stationery	268.79
Programs and abstracts (1948)	254.27
Roebling Medals (1948 and 1949)	279.15
New securities purchased	9,067.78
Commission on securities	74.14
Federal taxes on securities	8.50
Dividend and interest adjustments	30.86
Safety deposit box	7.80
Telegrams	4.02
Committee expenses	2.80
Society luncheon	7.10
Exchange charges on checks	1.50
Refunds	21.45
Travel expenses of officers to Annual Meeting	55.80
Back numbers of the Journal purchased	24.50
	$25,356.29
Cash balance, October 31, 1949	3,601.54
	$28,957.83

The endowment funds of the Society as of October 31, 1949, consist of the following securities:

Bonds

6M Atlantic Coast Line, 4½	$5,257.50	
5M New York Central, 5	4,300.00	
5M Southern Railway, 5	5,743.75	
4M Cleveland Union Terminal, 5	4,055.00	
4C Great Northern, 5½	400.00	
		$19,756.25

Preferred Stocks

200 shares, Southern California Edison, 4.88	$5,250.00	
100 shares, Union Pacific, 4	4,570.25	
60 shares, Jones and Laughlin, A, 5	4,987.50	
55 shares, United States Steel, 7	6,946.20	
50 shares, Virginia Electric & Power Co., 5	5,942.50	
24 shares, Public Service Electric & Gas Co.	728.40	
10 shares, Consolidated Edison, 5	1,066.64	
		$29,491.49

Common Stocks

240 shares, Potomac Electric Power Co.	$3,175.00	
60 shares, United Fruit Co.	3,067.50	
50 shares, Chesapeake and Ohio Railway	2,368.75	
50 shares, Pennsylvania Railroad	1,468.75	
41 shares, American Telephone and Telegraph	5,599.32	
40 shares, Kroger Co.	1,990.00	
28 shares, Standard Oil of New Jersey	1,444.84	
1 share, New York, Chicago & St. Louis Railroad	37.00	
		$19,151.16
		$68,398.90

Respectfully submitted,
Earl Ingerson, *Treasurer*

DANA FUND

Disbursements are made to needy mineralogists in war areas and to needy families of deceased mineralogists in war areas.

Receipts

Available balance, November 1, 1948	$372.42
Interest	8.42
	$380.84

DISBURSEMENTS

Disbursed	$ 40.00
Available balance, November 1, 1949	340.84
	$380.84

Respectfully submitted,
EARL INGERSON, *Treasurer*

REPORT OF THE AUDITING COMMITTEE

To the President of the Mineralogical Society of America:

The Auditing Committee has examined and verified the accounts of the Treasurer of the Mineralogical Society of America for the fiscal year beginning November 1, 1948, and ending October 31, 1949. The securities listed in the Treasurer's report, with all future coupons on the coupon bonds attached, are in the safety deposit box at the West End Branch of the Washington Loan and Trust Company in Washington, D. C.

Respectfully submitted,
FELIX CHAYES
JOHN C. RABBITT
GEORGE SWITZER, *Chairman*

ABSTRACTS OF PAPERS PRESENTED AT THE THIRTIETH ANNUAL MEETING OF THE MINERALOGICAL SOCIETY OF AMERICA, EL PASO, TEXAS, NOVEMBER 10–12, 1949

THE LEUCOXENE PROBLEM*

VICTOR T. ALLEN
Saint Louis University, St. Louis, Missouri

The author's interest in leucoxene began in 1934 during a study of the diaspore clays of Missouri. From 1939–46 leucoxene was observed in bauxite and high-alumina clays. *X*-ray patterns were made by P. F. Kerr, W. F. Bradley, J. W. Gruner, and J. M. Axelrod, and electron micrographs were contributed by the U. S. Bureau of Standards. The laboratories of the U. S. Geological Survey and the Illinois State Geological Survey made chemical analyses of new leucoxene samples that formed in the following ways: as a coating on ilmenite from Norway, as a clay derived from lavas from Hawaii, as a clay at Andersonville, Ga., as a pocket-filling clay at Roseland, Va. Hydrotitanite from Arkansas and xanthitane from North Carolina were also analyzed. These analyses served as controls for the petrographic and *x*-ray studies.

The following conclusions in regard to the nature of leucoxene resulted from these studies: (a) most of the opaque gray compound known to petrographers as leucoxene is amorphous; (b) the composition of leucoxene varies chemically and mineralogically; (c) *X*-ray patterns of leucoxene are similar to those of anatase, sphene, and rutile; (d) one per cent of well crystallized sphene may show in an *x*-ray pattern, so it is not known if some *x*-ray patterns represent the bulk of the leucoxene sample or a few well-crystallized grains; (e) a small amount of water is present in many analyses of leucoxene; whether this water is combined as a hydrate of titanium oxide or is adsorbed on the smallest particles of an oxide of titanium is not established; (f) leucoxene should be retained as a petrographic term for the alteration product in which titania occurs in rocks, but it must not be used as a mineral name implying a definite mineral species.

MICROSCOPE AND REFRACTOMETER FOR INFRA-RED LIGHT

RENE J. BAILLY AND KENNETH A. HOLKE
Washington University, Saint Louis, Missouri

In 1947, at the annual meeting in Ottawa, R. Bailly described a method of mineral determinations in infra-red light. In this method the eyepiece of the microscope or refractometer is replaced by a photoelectric ocular, and the variations of the photoelectric current, produced by the observed phenomena can be read on a very sensitive galvanometer. Generally a current amplification is needed. The method is rather long and requires a precise reasoning.

R. Bailly has improved a new method, based on the same principles with the use of an electronic image converter tube as the main part of the ocular instead of an electric photocell.

The invisible infra-red image produced by the lenses of the microscope or refractometer, is focused on the front, half-transparent cathode of the tube. Inside the converter, a set of electronic lenses bundles the beam of electrons coming from the cathode and focuses them on a fluorescent screen, producing a visible image of the observed field. This image, whose definition is very high, is magnified by an optical device. In this process, neither galvanometer nor amplifier is needed. The high voltage is supplied to the electronic lenses by a small power unit.

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* Published by permission of the Director, U. S. Geological Survey.

The use of the microscope and refractometer in infra-red light is exactly similar to their use for studies in visible light.

Amongst several researches, the variation of refractive index with the chemical composition is studied for the spinels and particularly for the family of chromite.

THE SYSTEM $NaAlSi_3O_8$-$KAlSi_3O_8$-H_2O

N. L. BOWEN AND O. F. TUTTLE
Geophysical Laboratory, Washington, D. C.

In the presence of water vapor under pressure soda feldspar and potash feldspar crystallize readily from their glasses. At high temperatures they form a complete series of solid solutions showing continuous variation of optical properties and lattice spacing as measured by *x*-rays. Studies of the dry melts had indicated that the solid solutions are of the type having a minimum-melting mixture. This relation is established beyond question upon investigation in the presence of water. At a pressure of 1000 kg/cm^2 of water vapor the minimum lies at the composition 70% $NaAlSi_3O_8$ and the temperature is 843°C. This temperature is 220° below that observed in dry melts, the lowering being induced by the presence of somewhat more than 7% H_2O in the liquid phase or melt. Increase of pressure to 2000 kg/cm^2 produces a further lowering of only about one-third as much. When crystallized at still lower temperatures the complete solid solution relation no longer obtains and two feldspars form side by side.

THE ALTERNATING LAYER SEQUENCE OF RECTORITE

W. F. BRADLEY
Illinois State Geological Survey, Urbana, Illinois

Rectorite is a complex layer type hydrous alumino-silicate related to the better known mica-like minerals. The structural scheme of rectorite consists of contiguous pairs of pyrophyllite-like units separated by pairs of layers of water molecules. An equally apt description would be the alternation of one pyrophyllite unit with one vermiculite unit. The nature of the arrangement is confirmed by the preparation of the equivalent complex in which ethylene glycol proxies for water.

GENERAL ASPECTS OF DISORDER IN MINERALS

M. J. BUERGER
Massachusetts Institute of Technology, Cambridge, Massachusetts

The order-disorder relation is a particular and non-classical variety of polymorphism. Polymorphic transformations are governed by the Helmholtz free energy relation, $A = E - TS$. It can be shown that E, which appears as the latent heat of transformation, is always positive, i.e., heat is absorbed with increasing temperature. In the classical polymorphic transformation, this occurs at a single temperature and coincides with the discontinuity of crystal structure at the transformation temperature. In disorder, there is no discontinuity, the heat being absorbed over a temperature interval culminating in the critical temperature of complete disorder. In this sequence, the basic crystal structure is not altered.

Disorder is ordinarily discussed for metals. Since metals have extremely simple crystal structures, only a simple and degenerate variety of disorder can occur in them. In more general crystal structures, disorder can occur in four distinct varieties: (1) Differential Interchange Disorder, (2) Interstitial Disorder, (3) Orientational Disorder, and (4) Distortional Disorder. Incidentally, distortional disorder is the mechanism of the high-low transformations in quartz, tridymite, and cristobalite.

The critical temperature of complete disorder falls with decreasing crystal size, and this accounts for the stabilization of high temperature forms at room temperature. The disorder transformation is accompanied by symmetry changes, the lower temperature form having the lower (subgroup, or derivative) symmetry. This is responsible for transformation twinning.

THE OPTICS OF TRICLINIC ADULARIA

URSULA CHAISSON
University of Chicago, Chicago, Illinois

Adularia has been generally believed to be monoclinic. Sections of adularia from several localities studied on the universal stage show optical orientations inconsistent with monoclinic symmetry. Sections normal to (001) show approximately 8° inclined extinction; parallel to (001), 6°; parallel to $(\bar{1}01)$, 15°. The optical deviation from monoclinic symmetry is absent or very small in the core of the crystals but increases toward the surfaces. In the (110) and $(\bar{1}\bar{1}0)$ sectors, inclination of extinction from the (010) symmetry plane is clockwise; in $(1\bar{1}0)$ and $(\bar{1}10)$ it is anticlockwise. Adularia from St. Gotthard and Tavetschtal, Switzerland, Mt. Vesuvius, Italy, and Valencia, Mexico conform to this pattern.

In monoclinic adularia, as has formerly been determined, nα lies in the symmetry plane and is very near to the a axis. In triclinic adularia the plane containing nα and the c axis has rotated about the c axis and forms an angle φ with the original symmetry plane (010). The following values of φ were measured on adularia from given localities:

St. Gotthard	12	different specimens
	10	
Tavetschtal	9	different specimens
	7	
Valencia	10	different specimens
Mt. Vesuvius	9	different specimens

The deviation of φ is clockwise in sectors (110) and $(\bar{1}\bar{1}0)$ anticlockwise in $(\bar{1}10)$ and $(1\bar{1}0)$.

Heating of triclinic adularia to 1000°C. for four days changed the optical orientation toward monoclinic symmetry.

EUDIALYTE AND EUCOLITE FROM SOUTHERN NEW MEXICO

STEPHEN E. CLABAUGH
University of Texas, Austin, Texas

Eudialyte and eucolite occur in Otero County, New Mexico, in small irregular dikes associated with the Wind Mountain laccolith, a conical mass of nepheline-analcime syenite porphyry about two miles in diameter. The laccolith is the largest of a group of Tertiary alkalic intrusives that make up the Cornudas Peaks of Texas and New Mexico located at the eastern margin of the Sierra Diablo Plateau about 50 miles east of El Paso. The eudialyte-bearing dikes are peripheral apophyses which penetrate short distances into contact metamorphosed limestones and shales of Permian and Pennsylvanian (?) age. Most of the eudialyte occurs in small subhedral to euhedral crystals about 0.5 to 3 mm in diameter; it is distributed irregularly in the dikes, and it is most abundant in narrow vein-like stringers that finger out into the calcareous rocks. The eudialyte ranges in color from rose pink and brown to colorless, and color is irregularly and zonally distributed in individual crystals. Eucolite occurs only as cores of some of the zoned crystals. A sample of eudialyte from the west side of Wind Mountain has the following indices of refraction:

$$\omega = 1.588 \pm .002$$
$$\epsilon = 1.593 \pm .002$$

The indices of different zones of some crystals differ by more than .005, and parts of the zoned crystals are essentially isotropic. The identity of the eudialyte was checked by quantitative spectrographic analysis. Aegirite, alkali feldspars, nepheline, analcime, apophyllite, zeolites, and contact metamorphic lime silicates are associated with the eudialyte.

Eudialyte has been reported from only two other localities in the United States, the Bearpaw Mountains, Mont., and Magnet Cove, Ark., but it is not uncommon as an accessory mineral in nepheline syenite pegmatite and related rocks in many other parts of the world, notably in the Fennoscandian region.

PETROLOGY OF THE RED RADIOACTIVE ZONES NORTH OF GOLDFIELDS SASKATCHEWAN

CHARLES E. B. CONYBEARE AND CHARLES D. CAMPBELL
The State College of Washington, Pullman, Washington

The red rocks that occur along faults and shear zones in the Precambrian granitic terrane north of Lake Athabaska are more radioactive than others in the area. They are mylonites ranging from protomylonite to ultramylonite, the latter generally forming anastomosing veinlets in the less intensely crushed types.

In the protomylonites the feldspar is deformed albite colored with hematite dust, especially along the cleavages; and unstrained minerals fill the interstices. In the more intensely crushed types the hematite dust permeates most of the rock; and the unstrained minerals, where present, form scattered aggregates or may locally replace the mylonite completely.

The first of these late minerals is clear albite, which forms rims on the red albite relicts. Then follow, in variable order and with repetitions, clear subhedral quartz and albite, specularite, and penninite containing radioactive anatase grains. The latest veins contain also pitchblende, calcite, and specularite. Oxidation products of pitchblende are locally abundant in solution cavities.

The early iron and later radioactive elements were introduced probably by solutions that rose along the mylonite zones. The other elements were probably present before mylonization; and all the late minerals, excepting pitchblende and calcite, may be products of recrystallization.

There is no evidence that the dusty hematite is an exsolution product of iron-rich feldspar.

SYNTHETIC QUARTZITE

H. W. FAIRBAIRN
Massachusetts Institute of Technology, Cambridge, Massachusetts

Quartz sand (grain size 125–250 microns) immersed in weak aqueous Na_2CO_3 solution is converted to sheared quartzite (grain size 100–200 microns) after a few hours' exposure to temperatures in the range 225°–450°C., confining pressures between 5,000 and 30,000 psi, and compressive loads between 35,000 and 115,000 psi. The principal feature of the experimental procedure is the use of a piston of rectangular cross-section and of a sand receptacle designed so that a biaxial strain is imposed on the quartz aggregate. The deformed prisms show shortening up to 60%, elongation up to 20%, and net decrease of volume up to 52%. Individual quartz grains have a maximum elongation of 2:1. Undulatory extinction bands are abundant, but no trace of deformation lamellae was found. There is little evidence of recrystallization. Both compaction and dimensional orientation are best developed in the upper range of temperatures and pressures used. Investigation of the lattice orientation (axis diagrams) of 35 deformed prisms shows random orientation for the most part. Calculation of the coefficient of correlation for each scatter diagram has also been carried out to test statistically this apparent randomness, but with inconclusive results.

Several similar experiments, using H_2O instead of Na_2CO_3, show a much lower degree of compaction and no trace of dimensional orientation; even, for example, after 6 days' exposure to 350°C., 30,000 psi confining pressure and 77,000 psi compressive load.

CONE-AXIS DIFFRACTION PATTERNS

D. JEROME FISHER
University of Chicago, Chicago, Illinois

Cone-axis *x*-ray pictures have been briefly described by Buerger (ASXRED Monogr. 1 1944). They are obtained when a lattice translation direction is given a precessing motion with respect to the direct beam, providing a flat film is placed in the holder normally occupied by the layer-line screen. The result is a series of diffracted spots which lie along concentric circles ("rings"), one for each level of the reciprocal lattice within range. A cone-axis photo bears about the same relation to a precession picture that a rotation film has to a Weissenberg.

While cone-axis photos are useful in determining the value of d^* in the precessing direction, they may also be employed to get diffraction symmetry and to check orientation; such uses will be illustrated. It is possible to index the spots of a cone-axis ring by a simple graphical procedure. This consists of making a trace-o-film enlargement of the spots to the proper scale to fit a precession photo of the same level. The trace-o-film is held in a framework on the arm of a universal drafting machine and its center is moved above a suitable guide circle drawn on the *n*-level precession photo. This recapitulates the actual motions of the precession instrument. Thus when a spot on the trace-o-film lies above a spot on the precession photo, the two have the same index, easily obtained from the precession picture. This technique will also be illustrated with simple cases that do not involve too many spots.

LIQUID IMMISCIBILITY IN THE Na_2O-SiO_2-H_2O SYSTEM.—PRELIMINARY DATA ON THE NA_2O-SiO_2-Al_2O_3-H_2O SYSTEM

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The system $Na_2O-SiO_2-H_2O$ was investigated at 400° and 450°C. The water rich immiscible phase present at 250°, 300° and 350°C. has a critical endpoint at about 400°C.

Some of the stability relationships of albite and analcite in the system $Na_2O-SiO_2-Al_2O_3-H_2O$ will be discussed.

SURVEY OF THE MINERALOGY OF URANIUM

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The descriptive mineralogy of uranium is discussed with regard to (1) The status of existing descriptions of the known uranium minerals, (2) The directions of needed investigation, and (3) The methods of identification and study. Particular reference will be made to the crystal structure of uranium minerals of the formula-type $A(UO_2)_2(XO_4)_2 \cdot nH_2O$ and to the correlation of optical, physical and chemical properties therewith.

DISORDER IN SULFIDES

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Heretofore metallurgists and physicists have made the most significant advances in the study of disorder, using for their examples intermetallic compounds. To use this knowledge to the fullest in initiating studies of disorder in minerals, sulfides were chosen, because of their submetallic character and because of the similarity between the covalent bonds in the sulfides and the metallic bond in the intermetallic compounds.

Bornite (Cu_5FeS_4), chalcopyrite ($CuFeS_2$), and stannite (Cu_2FeSnS_4) were chosen for laboratory study because these minerals are often found as the host minerals in exsolution textures. The presence of such textures is an indication that solid diffusion exists in the mineral at elevated temperatures. As all these minerals have two different atoms of similar size and properties (copper and iron), the chances of self-diffusion of these atoms in the structure and subsequent disorder when heat-treated would seem to be great.

X-ray, thermal, and electrical evidences indicate that bornite can exist in both a low and high temperature form, and that the structural difference between the forms is one of order-disorder. When thermally induced, the disorder first reaches a measurable magnitude at 170°C. and the critical temperature of complete disorder is reached at 220°C. By quenching in cold water, the high or disordered form can be retained, but if cooled slowly from the elevated temperature the low form is regained.

X-ray evidence indicates that both stannite and chalcopyrite can exist in a disordered form above a temperature of 600°C. However, as dissociation of these sulfides occurs below this temperature at atmospheric pressure, experimental technique has not yet been worked out to determine the thermal and electrical characteristics of the change.

THE SIGNIFICANCE OF GALLIUM AND GERMANIUM REPLACEMENTS IN SYNTHETIC FELDSPARS

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It is difficult to locate specific positions of Al and Si atoms in many silicates because of the approximately equivalent *x*-ray scattering power of these two atoms. In studying order-disorder relations in silicates, particularly in feldspars, location of these positions is desirable. The synthesis of feldspar structures in which Al^{+++} is replaced by Ga^{+++} and Si^{++++} by Ge^{++++}, and some of their crystal-chemical properties is discussed; the heavier elements make it possible to obtain more definite information on the degree of disorder. It is hoped that these gallium and germanium feldspars will help in the interpretation of some of the problems of the natural feldspars.

RHEOMORPHIC BRECCIAS

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Igneous intrusive breccias display the well-known features resulting from the injection, crystallization and reaction of an orthomagma. In contrast to these, many breccias in granitized areas contain undisturbed rock fragments and skialiths surrounded by granitic matrices which exhibit crystalloblastic textures and structures. These are considered to represent an intermediate stage of granitization and have been called replacement breccias.

Other plutonic breccias associated with granitization have crystalloblastic features similar to replacement breccias but differ from them by the presence of fluxion textures. At Cornucopia, Oregon, the matrices of some of these breccias show flow structures. At Sudbury, Ontario, some of the breccia dikes contain fragments which in part appear to merge into a crystalloblastic matrix having a pronounced flow structure. Other breccia dikes in this vicinity are filled with fragments of rocks of various kinds imbedded in a matrix replete with metamorphic minerals showing crystalloblastic features and marked flow alignment.

Breccias formed during metamorphism and exhibiting crystalloblastic features and flow structures cannot be adequately explained as orthomagmatic, but support the interpretation of movement of metamorphosed material. Since the mass flowage of this material can be included, but in a more restricted sense, under the broad definition of rheomorphism, it s suggested that breccias developed under such conditions be termed rheomorphic breccias.

ORBICULAR TINGUAITE DIKES NEAR BRYANT, SALINE COUNTY, ARKANSAS

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Tinguaite dikes probably of Cretaceous age have intruded the northwest edge of a 400 square mile province of phacolithic nepheline syenite intrusives in central Arkansas. The dikes are normal tinguaite and not metamorphosed Paleozoic pendants which also occur in this alkaline province.

The dikes vary in width from a few inches to 20 feet and occur in four parallel, linear zones spaced 200 to 500 feet apart. The zones are traceable for at least 2500 feet in a northeast-southwest direction and disappear beneath overlapping Tertiary sediments.

The dike magma was viscous and moved horizontally as well as vertically as evidenced by (1) linear flow lines along the unaltered wall rock contact (2) tortuous flow lines and eddy currents adjacent to wall rock protuberances and isolated, sparsely distributed, unaltered wall rock fragments and (3) cross-cutting flow lines analogous to schistosity in competent rocks. The wider dikes contain tabular phenocrysts of albite, while the narrower dikes contain small feldspathic spherulites.

The narrower dikes show unusually well developed orbicular structures heretofore not reported from alkaline rocks. The orbicules are 2 to 35 mm. in long dimension and consist of a tinguaitic nucleus and a peripheral banded zone about 1.5 mm. thick. In the peripheral zone bands of very fine grained aegirite alternate with bands of mixed nepheline, sodalite, potash and soda feldspars. There is no mineralogical difference between the orbicules and matrix, although there is a textural difference in that aegirite occurs as isolated aggregates in the matrix.

Contemporaneity of the orbicules with the late cooling stage is shown by their elongation in the direction of flow, denting of one by another, non-intergrowth, truncation by flow lines, and enclosure of flow lines.

BERYL AT MT. MICA, MAINE

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The discovery of gem tourmaline at Mt. Mica, Maine in 1820 was the beginning of nearly a century of gem mining at that locality. The last "pocket" opened in 1913 marked the end of active gem mining.

In June 1949, while mining feldspar a few hundred feet from the old gem workings, a new pocket was opened. This pocket is one of the largest found at Mt. Mica and measured about 20′×4′×5′. The lining of the pocket was mostly albite and muscovite, but a portion was of columnar beryl. Lying on the floor of the pocket were many colorless to white beryl crystals of unusual habit, completely terminated, and displaying many rare forms. Spectrographic analysis of these crystals shows the presence of approximately 1.0% lithium, 1.5% cesium and 0.1% rubidium.

THE ROLE OF YTTRIUM AND OTHER MINOR ELEMENTS IN THE GARNET GROUP

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Several minor and trace elements, notably yttrium, scandium and zinc are very common in garnets. The frequent occurrence of several of these in particular varieties of garnet suggests isomorphism. Yttrium, heretofore considered to be a rare constituent of garnets, is very prevalent in spessartites. It has been found to occur in concentrations of greater than 2 per cent Y_2O_3 in a few manganese-rich garnets. The frequent association of yttrium and manganese in spessartites suggests that ions of Y^{+3} with an ionic radius of 1.06 A have replaced ions of Mn^{+2} having an ionic radius of .91 A, the values being those of Goldschmidt. Scandium is most abundant in pyropes and Sc^{+3} with an ionic radius of .83 A may substitute for ions of divalent Mg (.78 A) or possibly for divalent Fe (.83 A) if sufficient almandite is present. Zinc is a common trace element in manganese, iron and magnesium-rich garnets and ions of divalent Zn (.83 A) may proxy for those of divalent iron. Other trace constituents detected in garnets include Ga, Ti, Na, Li, Dy, Gd, La, Ce, Nd, Pr, Sr, Ba, F, H_2O and Cb. Their hypothetical isomorphous relations to the major constituents are discussed. Included in the data are 7 new quantitative yttria determinations, visual spectroscopic analyses of more than 70 garnets and spectrographic analyses of 2 yttria precipitates obtained from spessartites.

HYDROXYL IN MINERALS

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Water may occur in minerals as mechanically held, evaporable "free" water, as loosely held "water of crystallization," and as more tenaciously held "water of constitution." Free water and tightly held hydroxyl (OH) may be differentiated in powdered minerals by their absorption of the infrared spectrum.

The OH group may be singly (monomeric) bonded to other elements in the crystal and therefore vibrate freely. Dimeric and polymeric bonding, where two or more OH groups share resonating mutual bonds, are indicated as also present by infrared absorption. Examples of the several bond types are illustrated by absorption spectrograms of clay minerals, opal, zeolites, and other hydrated minerals.

SURFACE AREA OF DEEP SEA SEDIMENTS

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The application of the radioactive inequilibrium method of age determination to unconsolidated ocean sediments is based on the preferential adsorption of ionium and radium ions on the surface of precipitating particles. The adsorptive properties of the sediment depend both on the chemical nature and the surface area of the particles. The chemical nature of the sediments can be ascertained from the mineralogy. For a constant mineral type the number of ions adsorbed will be proportional to the surface area. Therefore a knowledge of the mineralogy and the surface area of the material will permit the use of sediments of varying lithology in age determination by the radioactive inequilibrium method.

The surface areas of about 50 representative core samples have been obtained by the Emmett-Brunauer gas adsorption method. Values range from 2 square meters per gram for fine red silt to 40 sq. m./gm. for very fine inorganic calcium carbonate. Deep water red clay has a surface area of about 25 sq. m./gm. while the more common green clay from shallower water ranges from 10–15 sq. m./gm.

Implications of this quantitative information in age determination and ocean bottom geology will be discussed.

THE GEOMETRY OF TRICLINIC ADULARIA

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Thin sections of adularia show some optical deviations from monoclinic symmetry. These deviations are negligible at the core of the crystals, increase toward the surfaces, and reach a maximum in the area of the acute angle $(110) \wedge (\bar{1}10)$. *X*-ray pictures of this area show that the (110) and the $(\bar{1}10)$ sectors bear a symmetrical relationship to the plane (010) which is the symmetry plane in monoclinic adularia. The composition plane between these sectors is irregular in direction averaging close to (100) with an error of about 10°; it is always parallel to the *c* axis.

X-ray precession pictures of material from Mt. Vesuvius, Italy showed definite triclinic symmetry. Pictures were made of I, the (110) sector, and II, the $(\bar{1}10)$ sector. Precession axes were [100], $[\bar{1}00]$, [001], $[00\bar{1}]$, [101], $[\bar{1}0\bar{1}]$.

Results: (I) $\alpha=89°50'\pm5'$; $\beta=116°$; $\gamma=90°39'\pm5'$
(II) $\alpha=90°10'$; $\beta=116°$; $\gamma=89°21'$

The relationship between parts I and II fulfills no known law. They are, as stated above, simply symmetrical to the (010) symmetry plane of "monoclinic" adularia. This fact plus other considerations to be discussed imply that the adularia originally grew as a monoclinic crystal and later underwent a progressive change from the surface inward to a triclinic modification. *X*-ray as well as optical measurements of the core of the crystal with the triclinic system of coordinates given above showed no deviation from monoclinic symmetry.

Heating at 1000°C. for four days produced some optical change; the crystal became "less triclinic." The *x*-ray pictures still showed triclinic symmetry.

The combination of the optical and the *x*-ray measurements shows that the optical $n\alpha$ axis which is close to the geometrical *a* axis in monoclinic adularia deviates in triclinic adularia in the same direction as the geometrical *a* axis. The geometrical deviation is 39′ ($\gamma=90°39'$); the corresponding optical deviation is about 10°.

THE PHYSICAL ANALYSIS OF POLYCOMPONENT GARNET

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The widespread occurrence and compositional variation of garnet, in relation to the character of the rock facies in which it is indigenous, make it a useful diagnostic mineral in many petrologic problems. This places a premium on methods for the relatively rapid and accurate determination of garnet composition, especially where many specimens or very small amounts of clean material are involved. For detailed compositional correlation the 3-component solutions afforded by the triangular diagrams of Ford and Winchell do not suffice, since most garnet specimens (probably over 85%) contain 4 or 5 of the theoretical component "molecules" in significant amounts, that is, one mol per cent or more. Good 4-component solutions are afforded by the measurement of 3 physical properties which are independent (non-parallel) functions of the compositional variation, viz: index of refraction *n*, specific gravity G, and lattice constant a_0. These values may readily be applied to an algebraic solution or to the tetrahedral graphical solution of Philipsborn, and (with the same data) will yield quantitative results considerably better than the geometric estimates of Stockwell.

For most occurrences, however, only a 5-component solution in terms of pyrope, almandite, spessartite, grossularite, and andradite, will account for 99% or more of each garnet specimen. Such solutions are afforded by supplementing the 3 physical measurements (n, G, a_0) with a rapid partial chemical analysis for either MnO or FeO. With these 4 measured quantities, and the summation to 100% providing the fifth, it is possible to set up and solve 5 simultaneous equations to yield the values of the 5 unknowns, i.e. the molecular proportions of the 5 component molecules. In connection with a recent study of Adirondack garnets, a general solution of the 5 simultaneous equations has been made and the algebraic calculations thereby abbreviated to about 20 minutes of machine calculating. In laboratories having the usual equipment, an x-ray diffraction camera, thermally calibrated high-index liquids, a micro-pycnometer, etc., this composite method results in considerable saving of time over wet chemical methods, especially where many analyses are needed. By measuring N to $\pm.001$, G to $\pm.005$, a_0 to $\pm.002$ Å, and MnO to $\pm.2\%$, results accurate to ±1 mol per cent are obtained. Examples from the author's recent work are cited in illustration.

PETROLOGY OF BIG BEND NATIONAL PARK

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In the southern part of the Big Bend region, Texas, including the Big Bend National Park, occurs a great development of early Tertiary intrusive igneous rocks along with a thick section of pyroclastics and lava flows. The latter apparently once covered a large part of the area. Intrusive masses are mainly restricted to the southern part of the region but are abundant also in the Davis Mountains to the north.

The intrusive masses include a few larger plutons, stocks (?) and laccoliths emplaced under a cover of 3000–4000 feet of Tertiary and/or Cretaceous strata. There are many smaller laccoliths, sheets, sills and dikes.

The intrusive rocks are mainly fine grained with a few of coarser texture. Forty-two chemical analyses show a nearly complete gradation from silicic to mafic types. The suite is alkalic and generally sodic with phosphorus and titanium prominent in the intermediate and mafic members. These also exhibit considerable differentiation *in situ* with prominent formation of analcime. The silicic members do not exhibit marked differentiation but a considerable variation from mass to mass. It is possible that more than one sequence of intrusion is present. In the park area silicic sodic intrusives were late in the sequence.

The lava flows do not show the extensive variation of the intrusive rocks. They include however similar alkalic and other types and belong in the same suite. Vents have not been specifically determined but certain irregular plug-like masses in the southern Chisos Mountains probably were feeders for part of the rhyolitic lavas.

A PRELIMINARY ACCOUNT OF THE NATURALLY OCCURRING ANTIMONY OXIDES

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Among the naturally occurring antimony oxides the following phases have been recognized: senarmontite, Sb_2O_3, isometric; valentinite, Sb_2O_3, orthorhombic; and an isometric phase with the pyrochlore structure, and with the ideal composition $Sb^{III}Sb^{V}_2O_6(OH)$. The latter phase is by far the most common of the oxidation products of antimony minerals. Its composition and physical properties are exceedingly variable, due to the ready substitution of the antimony by other elements, and to the incomplete filling of some of the lattice positions. This appearance of vacant lattice positions is evidently largely responsible for the extreme variability of density and refractive index in this phase. Recorded values for the density range from 3.5 to 5.6, and of the refractive index from 1.6 to 2.1. This phase includes both the cervantite and stibiconite of the mineralogical texts. The phase Sb_2O_4 has not been recognized among the naturally occurring antimony oxides and its occurrence is questionable; the material that has been described as cervantite, with the supposed formula Sb_2O_4 (theoretical composition is Sb=79.2, O=20.8) is actually the above described isometric phase (theoretical composition for $Sb^{III}Sb^{V}_2O_6(OH)$ is Sb=76.4, O=21.8, H_2O=1.88).

NICKEL-COPPER-GOLD MINERALOGY AT THE MACKINAW MINE, SNOHOMISH COUNTY, WASHINGTON[*]

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A remarkable nickel deposit, the Mackinaw Mine, Snohomish County, Washington, is one of the few known in which the nickel sulfides and arsenides are not accompanied by pyrrhotite. The ore minerals are pentlandite, maucherite, niccolite, chalcopyrite and cubanite, magnetite, vallerite, sphalerite, and gold. There are two distinct types of ore, one containing pentlandite with little or no nickel arsenides or gold, the other, nickel arsenides and gold without pentlandite (or sphalerite). In many respects the mineralogy of this deposit resembles that of the Alistos mine, Sinaloa, Mexico, described by Krieger and Hagner in 1943 and then believed to be unique.

[*] Published by permission of the Director, U. S. Geological Survey.

PEROVSKITE FROM CALIFORNIA

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The first California find of perovskite is from the 910 level of the Commercial quarry at Crestmore, Riverside County. The mineral occurs as brilliantly shining, deep yellow octahedral crystals up to one mm. across. These are occasionally modified by small cube faces. Thin sections show the usual multiple twinning parallel to (100), and the individual lamellae are rather strongly birefringent. The perovskite is found in granular calcite, associated with numerous skeletal octahedral crystals of nearly black spinel, which in section is pale green in color.

HAUCHECORNITE

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A good small crystal of hauchecornite from the Friedrich mine, Hamm a.d. Sieg, Westphalia (Harvard Mineralogical Museum, 89710) is a thick square basal tablet *c*(001) combined with the tetragonal prisms *a*(010), *m*(110), and the tetragonal bipyramids *q*(011), *p*(021), *e*(111) ($a:c=1:0.7404$, goniometric and *x*-ray). Rotation and Weissenberg photographs around the *c*-axis and an *a*-axis show the Laue symmetry $4/mmm$, $a=7.28$ kX, $c=5.39$ kX ($\lambda CuK\alpha_1=1.5374$ kX), and the probable spacegroup $P4_22$. With the specific gravity 6.36 measured on this crystal (17 mg.) the four existing analyses on picked material indicate the cell content $Ni_8(Bi, Sb)_2S_8$. Strongest *x*-ray powder lines for identification: 2.79

kX (10), 2.39 (6), 2.30 (6), 4.34 (5), 1.861 (5). Hauchecornite is thus a well-defined mineral species.

PRELIMINARY REPORT ON THE SYSTEM LEUCITE-SiO_2-FeO

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A fairly complete reconnaissance has been made of this plane through the tetrahedron representing the more general system K_2O-Al_2O_3-SiO_2-FeO. An equilibrium diagram for the condensed system (each composition including Fe_2O_3 to the amount representing equilibrium with the pure iron crucibles) is given, along with a plot of the per cent Fe_2O_3 present at the liquidus temperature. The equilibrium diagram shows that the flat liquidus surface of fayalite covers a large portion of the system, it being the primary phase even in compositions very close to the binary potash feldspar-silica eutectic. The significance of these results in the crystallization-differentiation of natural magmas is discussed briefly.

GEOLOGY OF THE IRWIN DISTRICT OF COLORADO

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The Irwin District, west of Crested Butte, in Gunnison County, Colorado, is an area of geologic interest that goes beyond the local coal and silver deposits. The region has had a complex structural, stratigraphic, and intrusive history.

Through a thick series of sediments of varying ages and lithologies have come up a number of intrusive bodies, ranging from tremendous laccoliths to a swarm of dikes and sills.

This paper deals with some of the findings relating to the emplacement of the various igneous bodies. The deuteric effects, the structural deformation, the related hydrothermal deposits and the hydrothermal alteration are all discussed.

Intense epidotization of the sediments is a result of deuteric activity, but intensity is a function of type of igneous body, distance from contact and distance from original source of the igneous material.

Intrusion of the igneous material was apparently responsible for intense faulting and some folding. The faults became favorable loci of deposition of hydrothermal solutions which represented the end stages of igneous activity. The rich veins of arsenopyrite, native silver, gold, and ruby silver have made the area famous. The hydrothermal alteration of the igneous and sedimentary rocks is intense and has been studied by the writer.

DACITES FROM LAUGHLIN PEAK, COLFAX COUNTY, NEW MEXICO

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Laughlin Peak, one of 7 eroded volcanic cones of dacite in northeastern Colfax County, New Mexico, is located 11 miles north of Chico. In a previous paper the writer described specimens from this cone as vitrophyres, because of the large proportion of glass and lack of chemical data. Study of additional specimens and a chemical analysis show that the rocks from Laughlin Peak, the northeast side and top are similar to dacites on the other cones.

Hand specimens are pale-gray or gray and pink-banded, fine-grained, vesicular rocks. Micrometric analyses range as follows: Glass 83%–65%, plagioclase 21%–12%, hornblende

8%–1%, magnetite 14%–2%. The average mode of 7 slides is glass 73%, plagioclase 16%, hornblende 3%, magnetite 7%. The feldspar is oligoclase ($Ab_7 An_3$); hornblende usually the basaltic variety.

The norm (in part) calculated from the chemical analysis follows: Quartz 26%, orthoclase 15%, albite 36%, and anorthite 12%. Orthoclase slightly exceeds anorthite. Therefore the rock is close to the border between rhyolites and dacites, and could be called a soda-rhyolite or rhyodacite. The chemical analysis and norm are similar to those from Red Mountain and Cunningham Butte.

There is no marked difference chemically between these rocks. Although Laughlin Peak specimens are less crystallized, they do not differ mineralogically from the dacites of the other cones, Towndrow Peak and Red (or Bell) Mountain, both on Johnson Mesa, Cunningham Butte, Green Mountain, Raspberry Peak and Palo Blanco—the large cone 8 miles southeast of Laughlin Peak.

NONTRONITE AT BINGHAM, UTAH

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Nontronite has been found in abundance on the upper southwest levels of the Kennecott Copper Mine at Bingham, Utah. The rocks in this area were originally arkosic quartzites with dolomitic and/or argillaceous impurities. Contact action in these rocks has developed interstitial diopside, tremolite and pyrophyllite. Nontronite is present in localized areas as a weathering product of these minerals and orthoclase, where pyrite was present to supply iron for its development. Chemical analysis shows the mineral to have the formula

$$(\mathrm{Al}_{.59}\mathrm{Fe}_{1.02}\mathrm{Mg}_{.31})(\mathrm{Al}_{.52}\mathrm{Si}_{3.48})\mathrm{O}_{10}(\mathrm{OH})_{2\mathrm{x}.38}\,.$$

Differential thermal curves show a double endothermic peak between 600 and 700°C. as would be expected of a low iron nontronite.

ON THE CRYSTAL STRUCTURE OF BORNITE FROM ILLOGAN, CORNWALL

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A rotation photograph and a set of equi-inclination Weissenberg photographs were made of a single crystal of bornite from the Carn Brea Mine, Illogan, Cornwall, using filtered cobalt radiation. Although the three crystal axes are equal in length within the error of our determination, and the three interaxial angles are 90°, the structure is not truly cubic. The unit cell is pseudo-cubic with $a_0 = 32.8$ Å. The structure is approximated by the following arrangement in a cubic cell with $a_0' = 5.47$ Å ($= \frac{1}{6} \times 32.8$ Å): four copper or iron atoms distributed statistically over the four positions $\frac{1}{4}\frac{1}{4}\frac{1}{4}$, $\frac{3}{4}\frac{3}{4}\frac{1}{4}$, $\frac{1}{4}\frac{3}{4}\frac{3}{4}$, $\frac{3}{4}\frac{1}{4}\frac{3}{4}$, two copper or iron atoms distributed statistically over the four positions $\frac{1}{4}\frac{3}{4}\frac{1}{4}$, $\frac{3}{4}\frac{1}{4}\frac{1}{4}$, $\frac{1}{4}\frac{1}{4}\frac{3}{4}$, $\frac{3}{4}\frac{3}{4}\frac{3}{4}$, the total number of copper atoms in this cell being five and the total number of iron atoms being one on the average; four sulphur atoms on the positions 000, $\frac{1}{2}\frac{1}{2}0$, $0\frac{1}{2}\frac{1}{2}$, $\frac{1}{2}0\frac{1}{2}$. This approximate structure yields calculated intensities for all the strong and medium spots in good agreement with those observed; it yields small or zero calculated intensities for all the weak spots. The weak spots for which the calculated intensities are zero prove that the structure is actually more complicated, but the nature of the deviations from the approximate arrangement has not been found in our work.

ASSIMILATION (?) OF MICACEOUS SCHIST BY DIABASE

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Inclusions of highly altered micaceous schist in diabase were found by the author in the Umble Farm Quarry at Safe Harbor, Pa.

Rising through the magma these inclusions left a tubular trail. In cross section they show, megascopically, a rounded contour and are surrounded by a zone of hybrid rock of fairly uniform composition which sharply separates the inclusions from the diabase.

There is a close resemblance between this hybrid rock and some abnormal phases of the diabase formation exposed at other localities. Mineral and chemical compositions of the inclusions, the hybrid rock and the diabase are given.

No conclusions seem warranted but the occurrence suggests a possible origin for certain phases of the diabase intrusives.

HIGH-TEMPERATURE ALBITE

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Synthetic albite has optical and *x*-ray properties notably different from natural albite of granites and pegmatites, but like those of albite from some volcanic rocks. On prolonged heating at high temperatures pegmatitic albite inverts to a modification identical with the synthetic material. In the presence of fluxes the change takes place as low as 700°C. Albite twinning is observed in both forms.

MELONITE FROM BOULDER COUNTY, COLORADO

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Polished-section and *x*-ray studies of the telluride ores of Boulder County, Colorado, establish the fact that melonite ($NiTe_2$) in small amounts is widespread and is an early mineral in a complex epithermal sequence including a variety of sulfides, tellurides, and gangue minerals.

THE EFFECT OF POTASSIUM ON THE NEPHELINE-CARNEGIEITE TRANSFORMATION

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When the temperature of a melt of composition $NaAlSiO_4$ is lowered through its freezing point, crystals of carnegieite form. Carnegieite transforms sluggishly into nepheline at 1248°C. If one wishes to obtain nepheline directly from a melt this may be done by reducing the freezing point of the melt below this transformation point by the addition of a flux like lithium fluoride. Obtaining single crystals of nepheline by this method involves certain difficulties.

Recent crystal structure studies show that the composition $NaAlSiO_4$, while natural to carnegieite, is foreign to nepheline, and indeed is clearly forced on the crystals of nepheline by the above method of production. Normal nepheline requires one alkali atom in four to be potassium. In confirmation of this finding, naturally-occurring nepheline has a composition tending toward $KNa_3Al_4Si_4O_{16}$. Since the K atom is large for the available alkali sites in the carnegieite structure the composition $KNa_3Al_4Si_4O_{16}$ is foreign to carnegieite. It would appear possible to avoid the formation of carnegieite by cooling a melt of that composition.

Crystallization experiments show this line of reasoning to be correct. The x-ray powder pattern of the product obtained by cooling a melt of the composition $KNa_3Al_4Si_4O_{16}$ is found to agree with the pattern of natural nepheline.

This work was carried on in the Washken Laboratories, Cambridge, Massachusetts, under a development contract with the Squier Signal Laboratory, U. S. Army Signal Corps, Fort Monmouth, New Jersey.

GEOLOGY OF THE SILVER LAKE TALC AREA, SAN BERNARDINO COUNTY, CALIFORNIA

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The commercial talc-tremolite deposits near Silver Lake are within a persistent feldspar- and diopside-rich member of a highly metamorphosed pre-Cambrian sedimentary series. The deposits are elongate and lenticular and represent nearly complete replacement of carbonate strata. Other metasediments in the series include vitreous quartzite; marble containing disseminated forsterite, serpentine, chondrodite (?) and talc; quartz-muscovite schist; and quartz-biotite schist.

The quartz-biotite schist is commonly migmatitic; it also contains masses of microcline-quartz gneiss. An andesine-biotite-hornblende lamprophyre occurs as irregular bodies and dikes within the metasediments. Quartz diorite, which locally grades into and intrudes the lamprophyre, is the most extensive rock in the area. It intrudes the metasediments as sills and surrounds large residual metasedimentary islands. All of these metasedimentary and igneous rocks have been intruded by felsic dikes. The metasediments, in general, dip uniformly southward, as do planar structures in the gneiss, lamprophyre and quartz diorite.

The commercial deposits consist mostly of tremolite, but talc-concentrations are common near the walls and are apparently alterations of tremolite along shear zones. The tremolite rock contains local residual masses composed of earlier metamorphic minerals. Forsterite (?), the first of these early minerals to form, has been partly to wholly replaced by talc and tremolite of and early generation; calcite and serpentine have locally replaced this tremolite. Serpentine veinlets intricately traverse the residual masses but do not extend beyond their borders. The late-generation tremolite formed contemporaneously with the intrusion of felsic dikes.

THE JADEITE PROBLEM

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The jadeite problem is re-opened and new data are presented which redefine the probable stability range of jadeite. The mineral is known to occur in only three places; as small masses in serpentine with albite, or nepheline, or both, in Burma, and with albite and quartz in Japan. No specimens have been found in situ in the Central American occurrence. The interest in jadeite is mainly due to the theory that it is a high pressure mineral. If a reaction is known to be possible on other grounds, it will proceed under pressure in the direction of smaller volumes. However, the mere fact that the supposed products are of smaller volume than the reactants is not sufficient basis for inferring that the reaction will take place. All attempts to synthesize jadeite, even at pressures up to 4000 atmospheres, have been unsuccessful.

Although no direct structure determination appears to have been made, the common assumption that jadeite has a structure similar to diopside is justified by an indirect method. It is suggested that the coordination of sodium is probably less than normally assigned and that the "excess" aluminum occupies silica positions.

The temperature of formation is believed to be less that 800°C. since the mineral melts metastably at a temperature at least as low as 800°C.; it occurs with low temperature albite (<700°C.); it occurs in a reaction zone against serpentine (<500°C.); its chemical equivalents, nepheline+albite, are stable in the presence of water down to 600° and 3000 atmospheres; and its glass fails to crystallize dry below 800°.

A new analysis of jadeite and data on the diopside-jadeite and acmite-jadeite joins are given. The jadeite problem is considered to be distinct from the eclogite problem.

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1923 Washington, D. C.
1924 Ithaca, New York
1925 New Haven, Connecticut
1926 Madison, Wisconsin
1927 Cleveland, Ohio
1928 New York, N. Y.
1929 Washington, D. C.
1930 Toronto, Canada
1931 Tulsa, Oklahoma
1932 Cambridge, Massachusetts
1933 Chicago, Illinois
1934 Rochester, New York
1935 New York, N. Y.
1936 Cincinnati, Ohio
1937 Washington, D. C.
1938 New York, N. Y.
1939 Minneapolis, Minnesota
1940 Austin, Texas
1941 Boston, Massachusetts
1942 No meeting held
1943 No meeting held
1944 No meeting held
1945 Pittsburgh, Pennsylvania
1946 Chicago, Illinois
1947 Ottawa, Canada
1948 New York, N. Y.
1949 El Paso, Texas

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By vote of the Council of the Society, the list of subscribers is not printed this year.

MINERALOGICAL SOCIETY (LONDON)

A meeting of the Society was held on Thursday, November 10, 1949, in the apartments of the Geological Society of London, Burlington House, Piccadilly, W.1 (by kind permission).

The following papers were presented:

(1) Basaluminite and Hydrobasaluminite, two new Minerals from Northamptonshire.

By Professor S. E. Hollingworth and Dr. F. A. Bannister.

Fissures in the ironstone at Irchester, Northamptonshire, contain an assemblage of fine-grained minerals including two new hydrated aluminium sulphates, basaluminite $Al_4SO_4(OH)_{10}\cdot 5H_2O$ and hydrobasaluminite $Al_4SO_4(OH)_{10}\cdot 36H_2O$, together with allophane and halloysite. It is suggested that the concentration and precipitation of these minerals from originally dilute groundwater solutions followed on pronounced cambering of the strata during the Pleistocene period and that freezing to a depth of 60 feet played an essential part.

(2) An Account of the Antimony Mines of Great Britain and Ireland and of the Minerals found therein.

By Sir Arthur Russell.

(3) On Paratacamite and some Related Copper Chlorides.

By Dr. C. Frondel.

X-ray, optical, thermal and chemical measurements confirm the dimorphism of rhombohedral paratacamite with orthorhombic atacamite. The unit cell of paratacamite a 9.150 kX, α 96°28′ contains $8[Cu_2(OH)_3Cl]$. Both minerals have been identified in green patina and crusts of ancient copper and bronze objects, but paratacamite is the more common and is also formed by the action of warm sea-water on brass and copper. New measurements, including also *X*-ray powder data, are given for related minerals including tallingite, identified as paratacamite, and botallackite.

(4) Calc-silicate Skarn Veins in the Limestone of Lough Anure, Co. Donegal.

By Mr. W. S. Pitcher.

A series of calc-silicate skarn veins in a limestone mass surrounded by granite in the Dalradian of Western Donegal is described. The skarn bordering these veins exhibits a zonal arrangement of the constituent minerals idocrase, and garnet, wollastonite, diopside and tremolite enveloped by a zone of decolorized limestone; a central quartz-feldspar dykelet is present in some cases. The processes involved in the formation of the veins are discussed.

(5) The Chemical Composition and Physical Properties of the Residual Glass of the Kap Daussy Tholeiite Dyke, East Greenland.

By Mr. E. A. Vincent.

The fresh, unaltered residual glassy mesostasis of a Tertiary tholeiite dyke from East Greenland has been separated from the rock and chemically analysed. It has a somewhat unusual composition, containing 64% SiO_2 but being poorer in alkalies and richer in lime and iron oxides than is usual in igneous rocks with similar silica percentages. The relationships between refractive index, specific gravity and chemical composition are discussed, and comparisons made with the residual glass of the Kinkell tholeiite dyke (Walker, *Min. Mag.*, **24** (1935) pp. 131–159). The nature of this glassy residuum suggests that the dyke magma considered has not followed at all closely any of the well recognised trends of differentiation of basaltic magmas.

(6) Pisanite from Parys Mountain, Anglesey.

By Mr. Leslie Bor (communicated by Professor W. J. Pugh).

The paper describes pisanite ($(Fe,Cu)\ SO_4 \cdot 7H_2O$) as found at Parys Mountain, Anglesey, and gives the chemical and optical data of this mineral. It is thought that this records the first occurrence of pisanite as a British mineral.

(7) A Note on the Crystallography of Epidote.

By Mr. J. Hornstra and Prof. P. Terpstra (communicated by Dr. M. H. Hey).

In epidote, the zone axis [803] is almost perpendicular to the face (100), the departure being only $1\frac{1}{2}'$; accordingly, all those pairs of forms that are connected by the transformation determinant $\bar{4}0\bar{3}/040/004$ are goniometrically indistinguishable in twins on (100). Doubt is thrown on the occurrence of certain forms included in Matthes' list of established forms of epidote, and which might be well-known forms in twin position. The crystallography of a repeatedly-twinned epidote crystal from Sulzbachthal is described and figured.

(8) Dichroscopes for Microscope Stage and Ocular.

By Dr. A. T. J. Dollar.

The dichroscopes described are adapted for use on the stage or in the ocular of either a polarizing or non-polarizing microscope. They consist of groups of small plates of Polaroid, suitably oriented and mounted, which provide images of the absorption tints of a pleochroic mineral so interdigitated that small differences of hue, intensity of hue, or both, can be appreciated by direct ocular comparison.

(*Titles and abstracts kindly submitted by G. F. Claringbull, General Secretary.*)

At a meeting of the Mineralogical Society held on January 26, 1950, the following papers were presented.

(1) The Optical Properties and Composition of the Acmitic Pyroxenes.

By Mr. P. A. Sabine.

The chemical analysis and optical properties are presented of aegirine from a nepheline-syenite-pegmatite dyke forming part of the great post-Cambrian sill and dyke suit of Assynt, Sutherlandshire. The literature upon the acmitic pyroxenes has been reviewed and 20 of the published analyses recalculated to atomic proportions on the basis of $O+OH=6$. When plotted on a triangular diagram having $2(Na+K)$, $2(Mg+Fe^{+2}+Zr+Mn+Co)$ and $2(Fe^{+3}+V+Ti+Al)$ as vertices these analyses fall on the altitude from the $Mg+Fe+Zr+Mn+Ca$ vertex. Using this altitude as the abscissa of composition, curves are drawn for the refractive indices and optic axial angles. It is suggested that the definitions given previously for aegirine and aegirine-augite have been too rigid, and that a convenient division of the continuous series aegirine to aegirine-augite should be taken at $Na_{.45}Fe_{.45}^{+3}$, at or about which $a \wedge c=15°$ and $2V=90°$. Negative minerals would thus continue to be denoted as aegirine and positive ones as aegirine-augite.

(2) Si/Al Substitution in some Silicate Lattices.

By Mr. W. T. Harry (communicated by Dr. J. E. Richey).

Si/Al substitution in igneous clinopyroxenes, hornblendes, and biotites, varies within values characterising each group, greatest in biotite and least in the pyroxenes. In calciferous amphiboles Si/Al substitution increases from specimens in the Green schist metamorphic facies to those of magmatic origin. The data are discussed.

(3) THE AEGIRINE-GRANULITES OF GLEN LUI, BRAEMAR, ABERDEENSHIRE.

By Mr. G. R. McLachlan (communicated by Dr. S. O. Agrell).

The aegirine-granulites of Glen Lui are described. They are regarded as the results of metasomatism of garnetiferous mica-schists, the necessary soda being derived from a soda-pegmatite intruded into the overlying schists. Aegirine-augite, crossite and andradite are developed in the granulites with persistent relicts of almandine. These four minerals, the soda-pegmatite and two granulites were analysed.

(4) FURTHER DATA OF THE SLAG MINERALS NAGELSCHMIDTITE AND STEADITE.

By Mr. E. R. Segnit.

NOTES AND NEWS

CARNOTITE AND RADIOACTIVE SHALE IN MISSOURI*

GARRETT A MUILENBURG AND WALTER D. KELLER,
Missouri Geological Survey, Rolla, Missouri, and University of Missouri, Columbia, Missouri.

Carnotite and possibly other radioactive minerals have been found recently in limestone in Ste. Genevieve County, Missouri. This is the first reported occurrence of carnotite, not only in Missouri but in the Mississippi Valley region. It is of interest scientifically because it is one of the few known occurrences of this mineral in limestone as contrasted with the more usual occurrences in sandstone typical of the Colorado Plateau region. From the practical standpoint this discovery opens new areas, both geologically and geographically, to the search for radioactive materials. The possibility of finding commercially valuable radioactive mineral deposits in areas and under conditions hitherto not considered favorable will stimulate prospecting and may result in the discovery of other new sources of uranium.

The Ste. Genevieve occurrence was discovered in July, 1949, in a quarry in the Spergen limestone, about five and one-half miles north of Ste. Genevieve. Mr. Charles Bussen, the operator of the quarry, observed a thin film of a yellow substance along a joint in a block of limestone dislodged by blasting. This substance was identified as carnotite by the Missouri Geological Survey and Water Resources (Muilenburg). Investigations in the quarry by means of a Geiger-Müller Counter revealed a thin bed of black shale which is highly radioactive, separating two linestone beds. From its position in the section this shale parting is thought to be the source of the carnotite in the limestone below. The original identification of the mineral carnotite was subsequently confirmed by *x*-ray methods (Keller) and by spectrographic analysis (Dr. E. E. Pickett) at the University of Missouri laboratories.

Although carnotite is visible in only a few places in the shale, the entire mass is radioactive and there is a possibility that other radioactive minerals may be present in it.

An *x*-ray diffraction pattern of the black shale where no carnotite was visible indicates that the clay mineral is illite and the spectrographic analysis† shows that the shale contains uranium and vanadium. A tentative hypothesis therefore is that the illite contains these elements. Hendricks suggested that the vanadium of some of the western shales might be a part of the illite mineral.[1] Possibly the illite in the Ste. Genevieve shale may contain the uranium as well as the vanadium.

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* Published by permission of the State Geologist.

† Expenses for the *x*-ray and spectrographic analyses were defrayed under the Univ. of Mo. Research Council Grant No. 327 (302).

Further field and laboratory work on the occurrence, mineralogy, and origin of the radioactive minerals is being carried on under the direction of the authors.

SYNTHESIS OF UVAROVITE

F. A. Hummel, *The Pennsylvania State College,*
State College, Pennsylvania.

During the course of an investigation of pure compounds for use in ceramic fields, the synthesis of three garnets of the general formula $3CaO \cdot R_2O_3 \cdot 3SiO_2$ was attempted by dry reaction using chemically pure oxides as follows:

	Grossularite %	Andradite %	Uvarovite %
CaO	37.3	33.0	33.5
Al_2O_3	22.7	—	—
Fe_2O_3	—	31.5	—
Cr_2O_3	—	—	30.6
SiO_2	40.0	35.5	35.9

1. Grossularite

This mineral cannot be formed by dry reaction at atmospheric pressure. Extended heat treatments (150 hours) at 835, 1000, 1155, and 1240° C. all failed to produce grossularite, the principal reaction products being pseudowollastonite and anorthite. A glass of the grossularite composition yielded only pseudowollastonite and anorthite when devitrified. These results were to be expected in view of the previous work of H. S. Yoder (1), who emphasized the hydrothermal nature of the mineral.

2. Andradite

Extended heat treatments at 1105, 1200, and 1400° C. at atmospheric pressure failed to produce the mineral. The principal phase present was pseudowollastonite.

3. Uvarovite

The synthesis of pure $3CaO \cdot Cr_2O_3 \cdot 3SiO_2$ was achieved as low as 855° C. by holding for 150 hours. A 100 hour treatment at 1200° C. and a short duration run at 1400° C. also produced uvarovite. The x-ray data obtained by the use of a Norelco spectrometer ($CuK\alpha = 1.537\ k$X) are presented below:

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[1] Fischer, Richard P., Vanadium deposits of Colorado and Utah: *U. S. Geol. Survey, Bull.* **936-P,** 377 (1942).

Natural Uvarovite *Jacksonville, Calif.*		*Synthetic Uvarovite* 1400° C., 2 hrs.	
d	I/I_0	*d*	I/I_0
7.42	.1		
7.12	.2		
5.08	.23		
4.68	.34		
4.61	.27		
4.56	.26		
4.45	.26		
4.41	.26		
4.22	.26	4.20	.25
4.08	.19	4.14	.27
3.82	.14		
3.77	.14		
3.71	.13		
3.57	.2	3.20	.12
3.54	.17	3.18	.11
2.99	.53	2.96	.63
2.84	.14		
2.68	1.00	2.65	1.00
2.56	.16		
2.55	.16		
2.51	.14		
2.45	.76	2.43	.59
2.36	.25	2.33	.11
2.19	.23	2.17	.08
1.94	.35	1.96	.03
1.90	.2	1.93	.21
1.66	.37	1.66	.24
1.62	.18		
1.60	.98	1.60	.69
1.34	.28	1.34	.04
1.31	.31	1.31	.07
1.28	.22		

The natural uvarovite was carefully selected from a 1″×2″ specimen supplied by Hatfield Goudey of Yerington, Nevada. The linear thermal expansion coefficent of the synthetic product was found to be 85×10^{-7} cm/cm/° C. in the range 25 to 1000° C.

After heating to 1490° C. for one hour, the fine-grained synthetic green uvarovite changed to a coarse-grained gray-black material; *x*-ray and microscopic examination showed that the synthetic uvarovite had dissociated to pseudowollastonite and Cr_2O_3.

REFERENCE

1. YODER, HATTEN S., JR., Stability relationships of grossularite, presented at the twenty-eighth annual meeting of the Mineralogical Society of America at Ottawa, Canada, December 29–31, 1947. Abstracted in *Am. Mineral.*, **33**, March-April (1948).

INTERNATIONAL UNION OF CRYSTALLOGRAPHY
SECOND INTERNATIONAL CONGRESS AND GENERAL ASSEMBLY 1951

The Executive Committee of the International Union of Crystallography has accepted an invitation from the Swedish National Committee to hold the Second International Congress on Crystallography and the Second General Assembly of the Union in Stockholm from 27 June to 3 July 1951. These dates have been chosen in consultation with the Swedish National Committee and with the National Committees of all the countries which adhere to the Union. It is hoped that this early notice will make it possible for those interested in crystallography and its related fields to arrange to attend. Further information can be obtained from the General Secretary of the Union, Dr. R. C. Evans, Cavendish Laboratory, Cambridge, England.

Dr. Howard T. Evans who was in charge of the *X*-ray Diffraction Division of the Laboratory for Insulation Research at the Massachusetts Institute of Technology has joined the staff of the Philips Laboratories, Irvington-on-Hudson, New York. His work will be in the field of application of *x*-ray crystallography to the study of the physics of solids.

Norman Davidson, a geochemist of the U. S. Geological Survey, died December 31, 1949. He joined the staff as preparator in 1942. He continued his evening studies at George Washington University and was promoted to junior chemist. Successive promotions followed and in July 1949 he was sent to Denver as Head of the Investigations Section Analytical Laboratory.

Robert C. Vance, a long time employee of Ward's Natural Science Establishment, passed away in his sleep January 19, at Pittsford, N. Y.

Mr. Vance had been employed at Ward's Natural Science Establishment for 33 years, and was head of the mineral department from 1934 until the time of his retirement in November 1947.

He was a Fellow and Life Member of the Rochester Academy of Science and had served as Chairman of the Mineralogical Section of the Academy from 1936 to 1946. He was also a Fellow of the Society for Research on Meteorites, and a member of the Mineralogical Society of America, The Mineralogical Society of Great Britain and Ireland, The Walker Mineral Club of Toronto, and the Rocks and Minerals Association.

On January 1, 1950, the activities of the American Society for *X*-Ray and Electron Diffraction and the Crystallographic Society of America were officially ended, and a new society, the *American Crystallographic Association*, came into existence. This new society will carry on the activities of both the old ones. According to the aims stated in its new Constitution, "The object of this Society shall be to promote the study of the arrangement of atoms in matter, its causes, its nature and its consequences, and of the tools and methods used in such studies." The charter members of the new society totaling 496, have elected the officers for the first year, as follows: *President*, I. Fankuchen, Polytechnic Institute of Brooklyn; *Vice-President*, R. W. G. Wyckoff, National Institute of Health; *Secretary*, H. T. Evans, Jr., Philips Laboratories, Inc.; *Treasurer*, J. Karle, Naval Research Laboratories. Meetings will be held twice yearly, and the first one is scheduled for April 10–12 to be held at Pennsylvania State College. Further information concerning the American Crystallographic Association may be obtained from the Secretary, Dr. Howard T. Evans, Jr., Philips Laboratories, Inc., Irvington-on-Hudson, N. Y.

The Spring Meeting of the Society for Experimental Stress Analysis will be held at the Hotel Statler, Cleveland, Ohio, on May 25th, 26th, and 27th, 1950. Inquiries should be addressed to the Society for Experimental Stress Analysis, P.O. Box 168, Cambridge 39, Massachusetts.

Errata

The lazulite-scorzalite isomorphous series

W. T. Pecora and J. J. Fahey

Vol. 35, page 5, table 2: in analysis number 3, $TiO_2 = 0.20$ should read 0.22; in analysis number 4, total = 100.94 should read 100.40; in analysis number 6, add $ZnO = 0.17$.

Correction

In vol. 34, November-December, 1949, on page 838, lines 11 and 12, "a spectrographic analysis by K. J. Murata showed aluminum present in tenths of one per cent and calcium, zinc, and magnesium in hundredths," should have been written, "a spectrographic analysis by K. J. Murata showed Ti and Mg to be present in concentrations greater than 5 per cent, Fe between 1 and 5 per cent, Al and Mn in tenths of one per cent, Ca, Si, V, Zr, and Cb in hundredths of one per cent, and Cr and Sn in the thousandths."

Our sincere apologies to K. J. Murata.

(Signed) Joseph Murdoch
Joseph J. Fahey

BOOK REVIEWS

KRISTALLE UND GESTEINE (ein Lehrbuch der Kristallkunde und allgemeinen Mineralogie), by PENNTI ESKOLA, Wien, Springer-Verlag, 1946, 396+viii pp., 461 figs. Price about $11.00. First published (1939) in Finnish as *Kiteet ja kivet;* German translation by Martha Romer.

Here is a book that should be in the hands of every teacher of mineralogy. Of all books designed for an elementary course, this one is written from the most modern point of view. The treatment is sufficiently advanced to challenge and stimulate the intelligence of a college student, but without such rigorous prerequisites as vector analysis or physical chemistry.

The first section, *Crystal geometry* (76 pp.), is fairly conventional. The treatment of space lattices is good, and the fundamental subject of space-groups is given more than the usual cursory statement, although their relation to relative importance of surface forms is not mentioned. The crystal classes are logically discussed beginning with the triclinic.

Section II, *Crystal physics* (80 pp.), is modelled partly after Niggli's textbook.[1] Vectorial properties are given in terms of ellipsoids. Discussion of mechanical properties, although a little more extended than that in English texts, is still not commensurate with their importance in modern petrologic investigations. Crystal optics is treated at length in the manner of Tschermak-Becke, and Dana-Ford.[2] *X*-Ray methods are sufficiently covered in ten pages, and radioactivity is mentioned.

Eskola's treatment of *Crystal Chemistry* (80 pp.—enlarged from the Finnish edition) is his major contribution to mineralogy texts. Although it is of necessity based on original literature more than the other sections, it follows Evans' text[3] rather closely. Types of bonding are described from the standpoint of electron structure of the atoms. Ionic radii, polarization, and coordination are followed by a detailed discussion of crystal structures according to the classification of Evans. The eleven-page summary of *Geochemistry* might be read with profit by most geologists. Subject headings are: *Composition of the crust of the earth, Camouflage of the trace elements and their occurrence as independent compounds, Distribution and movement of the principal elements in the crust of the earth, Distribution of the elements in the concentric shells of the earth, Abundance of the elements, Origin of free oxygen in the atmosphere, and The Kuhn-Rittmann hypothesis of the origin of the earth from solar matter.*

As Eskola points out in the foreword, only a quarter of the book is devoted to "rocks." Under the heading *Physical chemistry of crystals. Rocks:* (70 pp.), he really presents "kinetic crystal chemistry." Elementary discussion of phase diagrams is followed by a review of processes of crystallization in igneous rocks; pegmatites are treated at length and related to hydrothermal solutions. A few pages are devoted to crystallization of chemical sediments. The section is weighted in favor of Eskola's principal interest, metamorphism, by a discussion only slightly condensed from his presentation in "Entstehung der Gesteine."[4] Throughout the section he attempts to discuss the subject as an application of mineralogy, and to this extent it prepares for, rather than replaces, a course in petrology.

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[1] Niggli, Paul, Lehrbuch der Mineralogie, 8th ed., Pt. 1, Berlin (1926).

[2] Becke, F., Tschermak's Lehrbuch der Mineralogie, 8th ed., Wien (1921); Ford, W. E., Dana's Textbook of Mineralogy, 4th ed., N. Y. (1933).

[3] Evans, R. C., Crystal Chemistry, Cambridge (1939).

[4] Barth, T. F. W., Correns, C. W., and Eskola, Pennti, Die Entstehung der Gesteine Berlin (1939).

In the final section (75 pp.) the common minerals are described on the basis of the combined chemical and structural classification of Strunz.[5] The discussion by groups is more interesting and helpful to the student than is the usual formal listing by species. Phase diagrams, structure drawings, and graphs and orientation diagrams of optical properties, are given for a few important cases.

If the book suffers at all it is in illustrations. Some of the diagrams are very good, such as the integrated series on the properties of the crystal classes. The scarcity of photographs may well have been dictated by economy. It is regrettable, however, that a drawing should be as noticeably in error as figure 303*a*, which purports to develop a body-centered lattice from close (square) packed layers.

It is fitting that one so eminently qualified and highly honored should publish an elementary text at the time that he is retiring from active teaching. His departure from tradition and convention in this text might well be followed by other authors, to the better training of all geologists in one of the fundamental branches of their science.

William T. Holser,
Dept. of Geology,
Cornell University,
Ithaca, New York

ROCK ALTERATION AS A GUIDE TO ORE—EAST TINTIC DISTRICT, UTAH.

By T. S. Lovering.

The first of the Monograph Series in Economic Geology is Rock Alteration as a Guide to Ore—East Tintic District, Utah, by T. S. Lovering, in collaboration with 11 other members of the U. S. Geological Survey. The choice of this study as the initial monograph publication is an auspicious one, for not only does the paper bid fair to become a classic description of the geology and mineralogy of an important active mining district, but also it outlines fundamental methods for the recognition, recording, and interpretation of wall rock alteration genetically related to ore deposits. The technique of detailed delineation of different zones of country rock alteration as clues to the presence of hidden ore promises to be of considerable value as an additional ore finding tool. As yet the method is in its teens, for too few geologists are acquainted with its requirements to allow its present widespread application. Furthermore its use requires considerable petrographic knowledge, especially of an unspectacular, humdrum group of clay minerals with whose optical characteristics all too few petrographers are familiar. Dr. Lovering is to be congratulated not merely on the geological quality of his product but also on his wise inclusion of mineralogical data that will assist others in the adoption of this device. Certainly the monograph will do much to advance the mapping of rock alteration zones in the examination of ore deposits and districts.

In the East Tintic District five definite stages of hydrothermal wall rock alteration can be recognized: (1) the early barren stage, (2) the mid barren stage, (3) the late barren stage, (4) the early productive stage, and (5) the productive (ore) stage. In stage one dolomitization of Paleozoic limestone and chloritization of the lower part of an Oligocene volcanic series were diagnostic changes. Stage two is characterized by "argillic" alteration products—endellite, beidellite, halloysite, kaolinite, dickite, and allophane. Minerals of the late barren stage are barite, pyrite, sparse chlorite, and SiO_2 as jasperoid in the sediments and quartz, allophane, barite, pyrite, and calcite in the lavas.

The introduction of potassium and the crystallization of clear quartz in open spaces characterize the early productive stage. In general during this period, earlier formed clay minerals were converted to sericite. During the last, or ore stage, pyrite and other sulfides were formed. In contrast to the often assumed idea that wall rock alteration and ore deposition proceed simultaneously, Lovering assigns the individual alteration zones to chemically ". . . differing solutions whose periods of activity have been separated by appreciable time intervals." The mineralogy of the zones changes outward in all directions from centers of hydrothermal activity. Outermost minerals are also the oldest; i.e., subsequent stages of alteration become increasingly restricted to the vicinity of solution conduits.

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[5] Strunz, Hugo, Mineralogische Tabellen, Leipzig (1941).

Superimposed on this complex sequential hydrothermal assemblage is a variety of supergene alteration products, including kaolinite, halloysite, jarosite, alunite, and allophane derived from acid sulfate waters, and montmorillonite, beidellite, endellite, allophane, gibbsite, and calcite formed from alkaline solutions. Recognition of different kinds of altered wall rock is facilitated by types of limonite whose diagnostic appearance depends upon the parent mineral—pyrite, biotite, chlorite, epidote, or magnetite.

The zones of alteration are related to the structure and lithology of the district by means of a series of maps and diagrammatic sections. An impressive frontispiece consists of a sectioned isometric block diagram in four colors showing relation of alteration to structure and ore in the North Lily fissure zone. The general mineral paragenesis is discussed in interesting detail, and the geochemistry of the alteration is summarized graphically from 37 new chemical analyses of fresh and altered rocks.

Lovering is careful to point out that the recognition of an altered zone is no sinecure for the presence of ore but states, "Knowledge of the space relations of altered zones and the stages represented in an area of favorable structure, and the position of the various stages in the sequence of hydrothermal events that culminated in ore deposition, gives a reasonably secure basis for estimating the chance of finding an ore body."

Successful application of the technique requires the integration of careful field mapping with detailed petrographic study—an exacting blend of the demarcation of regional features with the recognition of microscopic minutiae. Thus the attention of mineralogists and petrographers as well as that of economic and mining geologists may profitably be directed to this succinct summary of satisfying results obtained from a complex endeavor. In the 64 pages, 12 figures, and 5 plates contained in the paper-bound volume the author has competently formulated basic principles for the construction and utilization of a new weapon in the war on the diminution of ore reserves. The monograph may be obtained from M. M. Leighton, Business Manager, *Economic Geology*, Urbana, Illinois, for $1.50 to subscribers of *Economic Geology*, $1.00 to student subscribers of that journal, and $2.50 to non-subscribers.

E. Wm. Heinrich,
University of Michigan

INTERNAL STRUCTURE OF GRANITIC PEGMATITES. By E. N. Cameron, R. H. Jahns, A. H. McNair and L. R. Page.

As has been the case with many other geological inquiries, the study of pegmatites and their constitutents has passed in time through various phases, which have been distinguished one from another by changes in technique, by changes in approach and objectives, and especially by changes in emphasis on different aspects of the general problem. Early studies were concerned with pegmatites as sources of unusual mineral species and of fine crystals. A second phase, which began in the late 1800's and culminated in the early 1920's with the work of Landes, Schaller, Hess, and others, was marked by detailed attention to the sequence of mineral formation, focussed particularly on the less common and more intriguing mineral components. The third aspect, initiated just before World War II, found *geological* methods applied to pegmatite investigation in the first systematic attempt to decipher rules of their internal structure that might assist in finding and prospecting for mineable concentrations of pegmatite minerals. This phase has now been climaxed by the publication of Monograph 2 of the Economic Geology series—Internal Structure of Granitic Pegmatites. To Professor A. M. Bateman and his coworkers of the editorial staff of *Economic Geology* are due the universal congratulations and thanks of geological students for producing, in quick succession, two "smash hits" in the Monograph series. The same sentiments should likewise be extended to the authors, not only for their obviously painstaking compilation of the results of "68 man years of work in the pegmatite districts of New England, the Southeastern States, South Dakota, Idaho, Montana, Wyoming, Colorado, and New Mexico," but also for their definitive distillation of this gargantuan mass of diverse data.

The carefully organized Monograph of 115 pages is prolifically illustrated with 79 figures, including 7 collotype plates and 19 fold-in figures in the rear. Most of these are excellent and pertinent, and relatively few departures from the drafting quality appear throughout. The Monograph may be purchased from M. M. Leighton, Business Manager, *Economic Geology*, Urbana, Illinois, at a cost of $2.50 to subscribers of *Economic Geology*, $1.75 to student subscribers, and $4.00 to non-subscribers.

The introductory part of the work furnishes a background for the more specialized section by discussing some of the general features of granitic pegmatites, including distribution, age, relations to parent batholiths and to wall rocks, size, and shape. The largest single section is devoted, of course, to pegmatite structural units and describes first the zone types—border, wall, intermediate, and core with a detailed statement on the sequential petrology of zones. It is here, perhaps, that the single most significant contribution is presented—that despite individual and district variants, zoned granitic pegmatites show a relatively consistent petrological progression from walls to center, which is in accord with the demands of the theory of fractional crystallization of magmas. Thus this progression can be advanced as a powerful argument for the rather widely accepted theory, which is also favored by the authors ". . . that zones have developed from the walls inward, essentially by fractional crystallization and incomplete reaction in a restricted system."

The secondary units, which are fracture fillings and replacement bodies, are next described in interesting detail. However, the nadir of the work is revealed in the section devoted to the origin of these units (pp. 105–106). Here the authors content themselves with merely a weak-kneed rehash of the ideas advanced by previous pegmatite specialists. An adequate summary of the various hypotheses is disappointingly omitted. This is in direct contrast to the previous part on the origin of zones, in which the different theories are carefully weighed and a favorite is advanced. Surely considerable careful thought and much cogent argument have been expended on the genesis of secondary pegmatite units, and it is regrettable that the writers chose not to include a judicious inquiry into this aspect of the work and to present an equitable opinion. The reviewer believes, as do other workers in pegmatites, that this is one of the most fascinating and fruitful phases of the investigation and is now well removed beyond the bounds of mere specious speculation.

The Monograph concludes with a discussion of the practical applications of structural mapping of pegmatites and with a tag-along appendix on symbols suggested as suitable for depicting pegmatite units on maps, etc. The bibliography contains 121 entries. A most unfortunate omission of the study is any reference to the work of Scharizer (*Zeits. Kryst.*, **13**, 449–473, 1887), who not only at that early date specifically introduced the term, "zone" for pegmatite units ("Ich belege diese Mineralassociationen mit den Namen 'Zonen,' weil in der Natur in gewisser Hinsicht thatsächlich eine zonenartige Anordnung Platz greift . . ."), but also ascribed their development to magmatic differentiation in a closed system (". . . man im Pegmatitgranit von Schüttenhoffen sicherlich nicht das Werk zeitlich getrennter Bildungsepochen, wie etwa bei den Zonen der Erzgänge, vor sich hat, sondern diese Scheidung der Gesteinsbestandtheile in die drei angeführten typischen Associationen ist durch eine allmähliche und continuirliche Differenzirung eines ursprünglich einheitlichen Magmas bewerkstelligt worden"). Scharizer's pioneer and classic studies have long failed to receive the recognition they so richly merit. The Monograph might also have justly mentioned the work of Vlassov (*C.R. Dokl. Acad. Sci. U.R.S.S.* **41** (9), 384–387, 1943 and 53(9), 831–834, 1946) who has contributed significantly to the knowledge of internal pegmatite structure.

The publication marks a milestone in pegmatite study, and, indeed, some might wonder as to what remains for worthwhile future work. However, we are probably even now entering into the fourth phase of pegmatite investigations—that of correlating mineralogy, structure, and paragenesis with the geology of entire pegmatite districts. The summaries and definitions contained in this monograph are prerequisite to satisfactory essays along those lines in the future.

E. Wm. Heinrich,
University of Michigan

AN INTRODUCTION TO CRYSTALLOGRAPHY. By F. C. Phillips, pp. ix+302, 500 illustrations. Longmans Green and Company, New York, Price $6.50.

This is a very readable introduction to crystallography. The book is divided into two parts. Part I deals with the external symmetry of crystals and covers, in order, the fundamental laws of crystallography, methods of projection, drawing of crystals, a general survey of the crystal systems, goniometry, the thirty-two crystal classes, parallel growth and twinning, and the mathematical calculations of goniometry. The usage of the terms "diad,' "triad," "tetrad," and "hexad" axes will seem strange to many American crystallographers. The Weiss parameters, considered by many teachers of elementary crystallography to be very helpful to students in visualizing the orientation of crystal planes in space, are not included in this presentation. The discussion of projection and the drawing of crystals will be very useful. The chapter on goniometry is good, but it deals only with the one-circle method. Two-circle methods are dismissed in one sentence elsewhere in the book. The reviewer is greatly pleased with the treatment of the thirty-two crystal classes and the chapter on parallel growths and composite crystals. The chapter entitled "some mathematical relationships" will be greatly appreciated by students.

Part II, the symmetry of the internal arrangement, fills a need for an elementary presentation of the results of space group theory. The student who masters this section will be considerably helped in the study of *x*-ray crystallography. In this treatment of the crystal classes and of the space groups, the Hermann-Mauguin symbols are used. An appendix contains a brief discussion of the Schoenflies' symbols, which are introduced in order to enable the student to read papers employing that notation. The final chapter in the book concerns crystal habit and provides an introduction to the subject of morphological analysis developed by Professor Donnay.

The book is designed for mineralogists, chemists, physicists. and others. The illustrative examples include about 100 minerals and approximately 75 chemical substances. A human touch is added to this book by the inclusion of short biographical sketches of the lives of twenty-one crystallographers. The reviewer warmly recommends this book as a college text.

George T. Faust

NEW MINERAL NAMES

Zincfauserite

László Tokody, Cinkfauserite, uj ásvány Felsöbányáról (Zinkfauserit, ein neues Mineral von Felsöbánya): *Földtani Közlöny*, **79**, No. 1–4, 68–84 (in German 78–89) (1949).

Pale rose efflorescences from Level XII were collected and immediately sealed in glass tubes. Analysis by Elémer Schulek gave SO_3 33.54, MnO 19.14, ZnO 5.08, MgO 3.40, H_2O 39.61, Al_2O_3 0.11(?); total 100.88%. This corresponds to $MnO_{.65}MgO_{.20}ZnO_{.15}(SO_4) \cdot 5.25H_2O$. Crystallographic study showed the mineral to be a member of the epsomite group, orthorhombic with $a:b:c=0.9821:1:0.5615$. Forms observed: (100), (010), (101), (110), (111). Cleavage (010) good. Optically biaxial, negative, $2V$ large, $r>v$, n 1.465. Hardness 2½, D_4^{20} 1.9971. The mineral is readily soluble in water. It loses water rapidly in the air and is quickly covered by a white crust.

Discussion: Breithaupt in 1865 gave the name fauserite to an orthorhombic sulfate $(Mn, Mg)SO_4 \cdot 5.33H_2O$ with 19.61% MnO, 5.15% MgO. The validity of the species was questioned by Dana (1892) and has never been established. Tokody's description appears to be of a valid species, a member of the orthorhombic epsomite group from the crystallographic and optical data, though the sample had apparently lost water before analysis. However, the name zincfauserite is objectionable since zinc is a minor constituent. It would be better to transfer the name fauserite to Tokody's material.

Michael Fleischer

Csiklovaite

Sandor Koch and Julius Grasselly, Bismuth minerals in the Carpathian basin: *Acta Univ. Szeged., Sec. sci. nat., Acta mineral.*, petrog. **2**, 1–30 (in English 17–30) (1948).

Analysis by Grasselly of 0.2982 g. of homogeneous material gave Bi 67.76, Te 20.41, Se 1.37, S 9.97, Fe trace, impurities 0.40; total 99.91%. This gives Bi:Te:S+Se=2.02:1:2.04, or $Bi_2Te(S, Se)_2$. The mineral is very similar to tetradymite, but differs in that in polished section it is gray, lacking the creamy tinge of tetradymite. In oil immersion it is anisotropic, light to dark bluish gray, tetradymite is more pinkish gray. More rapidly etched by 1+1 HNO_3 than tetradymite. From Csiklova (Ciclova), Roumania.

M. F.

Stannopalladinite

I. N. Maslenitzky, P. V. Faleev and E. V. Iskyul, Tin-bearing minerals of the platinum group in sulfide copper-nickel ores (in Russian): *Doklady Akad. Sci. USSR*, **58**, 1137–1140 (1947); through *Mineralog. Abs.*, **10**, 453 (1949).

Analysis of grains separated from these ores (presumably from Norilsk) are given. Stannopalladinite, as elongated and rounded cubic crystals closely associated with the more abundant magnetic ferroplatinum, is non-magnetic. Analysis gave Pt 15–20, Pd 40–45, Fe 0.3–2.3, Sn 28–33, Cu 5–12, Ni 0.1–0.7, Insol. 0.25–2.5%; corresponding to Pd_3Sn_2. Analyses are also given of ferro-platinum and of an unidentified mineral which contained Pt 51–63, Pd 12–20, Sn 21–22, Fe 0.6–0.9, Cu 0.3–2.2, Ni 0.45, Insol. 1.0–2.3%.

M. F.

Asbophite

F. V. Syromyatnikov, Asbophite—a new variety of serpentine (in Russian): *D. S. Belyankin Jubilee Vol., Acad. Sci. U.S.S.R.* **1946**, pp. 32–40; through *Mineralog. Abs.*, **10**, 454 (1949).

"Material from the serpentine-asbestos deposit of Bazhenovo, northern Urals, has alpha=1.559, beta=1.562, gamma=1.570, $2V=64°$, positive; the x-ray Lauegram differs from that of chrysotile. Analysis gave: SiO_2 40.01, TiO_2 trace, Al_2O_3 0.79, Cr_2O_3 trace. Fe_2O_3 1.87, FeO 0.42, NiO 0.12, MnO 0.08, MgO 39.22 CaO 2.06, $(Na, K)_2O$ 0.09, S 0.02, H_2O+12.06, H_2O-0.67, CO_2 2.92, volatile material 0.12; total 100.45%."

M. F.

Paragearksutite

N. A. Smolyaninov and E. N. Isakov, New alumocalcium fluorides (in Russian): *D. S. Belyankin Jubilee Vol., Acad. Sci. U.S.S.R.* **1946,** pp. 145–151; through *Mineralog. Abs.*, **10,** 453 (1949).

"White massive material (*n* 1.454) occurs with kaolin in cavities in quartz veins at the wolframite deposits in Transbaikalia. The mean of three closely agreeing analyses, F 35.25, Al_2O_3 31.08, Fe_2O_3 0.56, CaO 30.03, SiO_2 1.56, H_2O+16.19, $H_2O-0.59=115.26$, less O for $F=100.42\%$, gives the formula $4CaF_2 \cdot 4Al(F, OH)_3 \cdot 3H_2O$, which differs from that of gearksutite in containing slightly less water, and the mineral is named paragearksutite. Another analysis after deducting 81.7% kaolin and 3.3% limonite gives a formula $2CaF_2 \cdot 4Al(F, OH)_3 \cdot 3H_2O$."

M. F.

Illidromica

Ciro Andreatta, New hydrothermal member of illite-hydromica series; transitions among mica, hydromica, and montmorillonite (in Italian): *Periodico Mineral.* (*Roma*) **18,** 11–31 (1949); through *Chem. Abstracts*, **43,** 7871–7872 (1949).

Illidromica forms a layer 1–2 m. thick, below halloysite and montmorillonite clays. Analysis gave (after deduction of rutile (about 1%), kaolin (about 5%), pyrite, and 6.8% H_2O-): SiO_2 53.12, Al_2O_3 27.36, Fe_2O_3 2.64, FeO 1.06, MnO 0.07, MgO 2.62, CaO 0.53, K_2O 3.51, Na_2O 0.49, H_2O+8.60%; formula $K_2Mg_2Al_{12}Si_{20}O_{55}(OH)_{12} \cdot 4H_2O$. The mineral contains mixed illite and montmorillonite layers. Optically negative, with alpha = 1.556–1.558, gamma = 1.583–1.608, $2V$ 0–20°, rarely up to 35°.

M. F.

Pulszkyite

Jozsef Krenner, Pulszkyit, új magyar ásvány (Pulszkyite, a new Hungarian mineral): *Földtani Közlöny*, **78,** 205–206 (1948) (Hungarian and German).

This note by L. Tokody gives the data found in Krenner's notebook after the latter's death. Krenner described the mineral at a meeting in January, 1915, but never published the data. The mineral is apple-to grass green, found in hexagonal plates with (001) dominant, optically uniaxial, negative. It contains Cu, Mg, and SO_3, hence is a copper magnesium sulfate. The mineral occurred with herrengrundite at Úrvölgy, Hungary (now in Slovakia). Study of 27 specimens from Úrvölgy in the Museum Collection failed to locate any with this mineral. The name is for Ferenc Pulszky, 1814–1897, Director of the Hungarian National Museum.

Discussion: The rescue of this name from oblivion was no service to mineralogy.

M. F.

Montesite

Roberto Herzenberg, Montesita, nuevo mineral de estaño en Bolivia: *Mineria Boliviana*, **6,** No. 44, 5–7 (1949).

Three analyses are given of supposed teallites. That from Carguayco Ilo corresponds to teallite, that from Monserrat to zincian teallite (Zn 14.1%); the third, from Lípez Huayco-Ocurí, gave Ag 0.52, Sn 52.4, Sb 0.80, Pb 23.8, Zn not determined, Fe 0.46, S 18, 7; insol. 0.90, 0 (calcd.) 1.10, (cassiterite 5.2%). The analysis was made on carefully selected material, which did contain some cassiterite. (No statement is given as to how the cassiterite content was determined. M. F.). The analysis corresponds to $PbSn_4S_5$, which is considered part of the isotypic series between teallite ($PbSnS_2$) and herzenbergite (SnS).

The mineral is silvery-white, hardness above $2\frac{1}{2}$, but less than 3, does not mark paper; sp. gr. 5.66.

The name is for Don Ismael Montes, twice president of Bolivia and founder of the Escuela Práctica de Mineriá at Oruro (1905).

Discussion: This cannot be accepted as a valid mineral species pending proof by optical and *x*-ray study of the homogeneity of the material.

M. F.

Unnamed

M. Fornaseri, Un nuovo ossicloruro di antimonio fra i prodotti di alterazione dell' antimonite alle Cetine di Cotorniano presso Rosia (Siena): *Atti* (*Rend.*) *Acad. Nazionale Lincei, Classe sci. fis. mat. e nat.*, Ser. 8, Vol. **3**, 365–369 (1947).

Analysis of white, fibrous crystals occurring as an alteration product of stibnite gave: Sb 78.6, Cl 7.24, H_2O none, O (by difference) 14.16; corresponding to Sb_3O_4Cl or $4Sb_2O_3 \cdot SbCl_3$. Goniometric measurements, difficult because the crystals were distorted and striated, gave $a:b:c=1.065:1:?$, α 129°26′, β 138°, γ 73°9′; triclinic. Forms observed were (010), (110), $(1\bar{1}0)$, and (001). The *x*-ray pattern differs from those of valentinite and hydroromeite. Optically negative; elongation positive. Easily soluble in cold concentrated HCl, soluble, but less easily, in NaOH. Heated in the closed tube gives oily droplets of $SbCl_3$ and a sublimate of minute octahedra of Sb_2O_3. Differs from the three synthetic antimony oxychlorides described in the literature.

M. F.

Scorzalite, Souzalite

W. T. Pecora and J. J. Fahey, *Am. Mineral.*, **34,** 83–93 (1949).

Sengierite

J. F. Vaes and P. F. Kerr, *Am. Mineral.*, **34,** 109–120 (1949).

Retgersite

Clifford Frondel and Charles Palache, *Am. Mineral.*, **34,** 188–194 (1949).

Bayleyite, Andersonite, Swartzite

Joseph Axelrod, Frank Grimaldi, Charles Milton, and K. J. Murata. *Am. Mineral.*, **34,** 274 (1949) (abs.).

Rockbridgeite, Laubmannite

Clifford Frondel, *Am. Mineral.*, **34,** 513–540 (1949)

Frondelite

M. L. Lindberg, *Am. Mineral.*, **34,** 541–549 (1949).

Torreyite (formerly delta-mooreite)

Joan Prewitt-Hopkins, *Am. Mineral.*, **34,** 589–595 (1949).

Wolfeite

Clifford Frondel, *Am. Mineral.*, **34,** 692–705 (1949).

DISCREDITED MINERALS

Warrenite (=Owyheeite)

S. C. Robinson, *Am. Mineral.*, **34,** 398–402 (1949).

Goongarrite (=Cosalite+Galena)

R. M. Thompson, *Am. Mineral.*, **34,** 459–460 (1949).

Warthaite (=Cosalite)

R. M. Thompson, *Am. Mineral.*, **34,** 459–460 (1949).

Sjögrenite (of Quensel)=Dufrenite

Clifford Frondel, *Am. Mineral.*, **34,** 521–523 (1949).

It is shown that the mineral name sjögrenite by Krenner (see *Am. Mineral.*, **32,** 484 (1947)) is dufrenite, and the name sjögrenite should be restricted to the member of the hydrotalcite group.

Bodenbenderite (=Spessartite garnet)

Charles Milton and A. T. Myers, *Am. Mineral.*, **34,** 608–611 (1949).

M. F.

REQUEST FOR EARLY ABSTRACTS FOR ANNUAL MEETING OF MINERALOGICAL SOCIETY OF AMERICA IN WASHINGTON, D. C., NOVEMBER 16–18, 1950

Although the deadline for abstracts remains September 1, it is requested that as many abstracts as possible be in the hands of the Secretary by June 1. Because of the increasing number of papers that are submitted for oral presentation at the annual meetings, it will probably be impossible to include on the program every one who submits a paper. Thus, the type of sessions will be based on the papers for which abstracts have been received by June 1; and the papers, for which abstracts are received later, will be fitted, if possible, into the established framework.

C. S. Hurlbut, Jr., *Secretary*